Nuclear Magnetic Moments of Nitrogen^{14}, Chlorine^{37}, and Indium^{115}

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NUCLEAR MAGNETIC MOMENTS OF NITROGEN^{14}, CHLORINE^{37}, AND INDIUM^{115}

Abstract of

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NUCLEAR MAGNETIC MOMENTS OF NITROGEN^{14}, CHLORINE^{37}, AND INDIUM^{115}

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B.S., United States Naval Academy, 1934

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The "extra-nuclear" properties of the nucleus are discussed and the importance of accurate measurements of these properties in testing future nuclear theories is pointed out. The various methods used to measure one of these properties, the nuclear magnetic moment, are described.

Nuclear magnetic moments μ_I of Nitrogen^{14}, Chlorine^{37}, and Indium^{115} were measured by means of a magnetic resonance spectrometer of the super-regenerative type. Improvements of the modulation methods and of the magnetic field homogeneity of the spectrometer previously employed are discussed.

The values obtained for the various μ_I were

$$\mu_I(N^{14}) \qquad (0.40361 \pm 0.00002)\mu_n,$$
$$\mu_I(Cl^{37}) \qquad (0.683722 \pm 0.000048)\mu_n, \qquad \text{and}$$
$$\mu_I(In^{115}) \qquad (5.50945 \pm 0.00015)\mu_n.$$

These results are observed to compare satisfactorily with those previously obtained by investigators at Stanford using another type of magnetic resonance spectrometer. Results obtained by these methods in the cases of In^{113} and In^{115} are seen not to agree with those obtained by the atomic beam method. An explanation offered

19389

by Foley for this lack of agreement is summarized.

The results are further compared with those predicted by the Schmidt equations and by using in these same equations the value of the angular momenta of the odd proton for Cl^{37} and In^{115} obtained from nuclear shell theory. A discussion of the relationships between μ_I and the nuclear quadrupole moment Q is also given with the conclusion being reached that to date no theory has yet been successful in establishing any important relationships between these two quantities.

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TABLE OF CONTENTS

Page
I. Introduction 1
(a) Nuclear Mass 3
(b) Nuclear Spin 3
(c) Nuclear Magnetic Moments 5
(d) Nuclear Quadrupole Moments 8

II. Methods of Measuring Magnetic Moments

(a) Hyperfine Structure 11
(b) Molecular Beams 13
(c) Work on Liquids and Solids 18
(1) Method of Purcell and Pound . . . 24
(2) Method of Bloch 29
(3) Super-regenerative Method . . . 32
(4) Heterodyne Method 38

III. Experimental Arrangements

(a) Apparatus 43
(b) Modulation Improvements 46
(c) Homogenizing of the Magnetic Field . . . 47
IV. Experimental Results 58
(a) Indium^{115} 59
(b) Chlorine^{37} 60
(c) Nitrogen^{14} 61
V. Discussion of Results 62
(a) Shell Theory 65
(b) Relationships Between μ_I and Q 69

19349

I. INTRODUCTION

The present status of the theory of the atomic nucleus is roughly comparable to that of the extra-nuclear atom circa 1912, by which time a mass of spectroscopic and x-ray data had been assembled and a number of empirical relationships established, yet a satisfactory model of the electronic configuration had not been proposed. However, within the ensuing few decades this wealth of data sufficed to produce first the Bohr model of the hydrogen atom and later the Lande vector model, the concept of closed electronic shells and the introduction of electron and nuclear spins, all of which contributed greatly to the explanation in classical or semi-classical concepts of a great many of the phenomena observed involving bound electrons. The later of these developments were more or less paralleled by de Broglie's proposal of the wave nature of matter in 1925 and the development of matrix mechanics and wave mechanics by Heisenberg and Schroedinger, respectively, within the following one or two years.

The status of the theory of the nuclear atom is today quite similar to the above situation. The data assembled by a host of experimenters in recent years relative to nuclear energy levels has, particularly with the aid of wave mechanics, enabled physicists to offer a certain number of fairly plausible explanations of basic nuclear phenomena. Even so, many other basic phenomena are not yet adequately explained and no theory yet proposed appears to hold forth very great expectations. One of the new attempts to systematize the prediction of nuclear properties is the fairly recently proposed theory of closed shells in nuclei, but this theory as yet has produced certainly no

more than a limited number of satisfactory predictions of nuclear properties.

One of the great handicaps suffered by the nuclear physicist is the difficulty he encounters in trying to obtain accurate data pertaining to nuclear energy levels. Fairly precise measurement of alpha particle energies is difficult, but really accurate measurement of beta particle, gamma ray, and neutron energies presents even greater problems, whereas the measurement of many of the lower energy phenomena is now, perhaps, impossible. Indeed, in many cases the experimenter is greatly handicapped by not being able to identify accurately the nuclide from which phenomena involving radioactivity originate.

In contrast to this general cloud of less than moderate accuracy which surrounds quantitative measurements of nuclear phenomena, there are certain observable quantities of the nucleus referred to frequently as "extra-nuclear properties" which are capable of being measured with remarkable precision using techniques now available. These are the properties of nuclear charge, nuclear mass, nuclear spin, nuclear magnetic moment and, to a less accurate degree of measurement, the nuclear quadrupole moment. The nuclear charge for those elements occurring naturally on earth has already been completely determined in terms of the electronic charge. It may reasonably be expected that any satisfactory theory of the nucleus must account adequately and accurately for all of these extra-nuclear properties as a first test because of the superior accuracy of data assembled or being assembled relative to these properties.

(a) Nuclear Mass

Absolute measurements of nuclear masses are now limited by the accuracy with which Avogadro's number has been determined. However, relative mass determinations made with the mass spectrograph may be obtained accurate to one part in fifteen thousand in the case of light atoms and to one part in ten thousand for atoms of greater mass number.[1] This accuracy is sufficient to measure mass changes in all but very low energy nuclear reactions involving emission of gamma rays. By the use of Einstein's formula for the equivalence of mass and energy one can from mass spectrographic data obtain the energies involved in a great number of nuclear reactions.

(b) Nuclear Spin

Nuclear spin was first proposed by Pauli in 1924 to explain hyperfine structures (excluding isotopic effect) by assuming that the nucleus possessed a magnetic moment which perturbed the motion of the orbital electrons.[2] Not until 1926, however, when Goudsmit and Uhlenbeck gave adequate explanation of the possibilities of accounting for fine structure in atomic spectra by assigning to the electron

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[1] Halliday, D. *Introductory Nuclear Physics*. New York: John Wiley & Sons, 1950, p. 258.

[2] Pauli, W. *Naturwiss* *12*, 741 (1924).

an inherent spin* of $\frac{1}{2}\left(\frac{h}{2\pi}\right)$ did it become evident that nuclear spin might indeed explain certain hyperfine structures. Using the concept of space quantization of the nuclear spin vector together with the vector model of the extra-nuclear atom one could ascertain that the inherent part of the nuclear angular momentum might be determined merely from the number of hyperfine components in a spectral line. This inherent or spin angular momentum of the nucleus is given by $I\left(\frac{h}{2\pi}\right)$, where I is called the "nuclear spin" and has zero, integral or half-integral values. For a given nucleus in the ground state I has a fixed value.*

Another method of determining nuclear spins is afforded by observing the optical spectra of diatomic molecules containing identical nuclei, for in such spectra there is an alternation of intensity in the band spectra in the ratio of (I+1)/(I). Thus the measurement of relative intensity of alternate lines in band spectra of certain diatomic molecules can be used to determine nuclear spin.

The investigation of nuclear spins has revealed that spins of the elementary nuclear particles, neutrons and protons, in the ground state have spin angular momentum values of $\frac{1}{2}\left(\frac{h}{2\pi}\right)$. Furthermore the

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*The lengths of the nuclear spin and the electronic spin angular momentum vectors are shown by the wave mechanics to have the magnitudes $\sqrt{I(I+1)}$ and $\sqrt{s(s+1)}$, respectively, in units of $\left(\frac{h}{2\pi}\right)$. However, the maximum projected values they may possess in any specified direction, say, that determined by an external magnetic field, are I and s.

Pauli exclusion principle applies in the case of these individual elementary particles of a nucleus as well as for the bound atomic electrons. It has also been determined empirically that a nucleus with an even number of nucleons will have the values I = 0, 1, 2,····, whereas one with odd mass number will have I = 1/2, 3/2, 5/2,···. In so far as present day observations have revealed it may also be said that nuclei having an even number of both protons and neutrons have, at least in their ground states, zero resultant angular momenta and zero nuclear magnetic moments.

(c) Nuclear Magnetic Moment

According to classical electromagnetic theory the magnetic moment of a spinning charge cloud with mass m and charge e uniformly distributed throughout its mass should be $\frac{e}{2mc}\cdot p_s$, where p_s is the angular momentum due to spin. This classical theory is not borne out by experiment, which reveals a magnetic moment of value $2\cdot\frac{e}{2mc}\cdot p_s$ for the electron due to its spin, which is just twice the classical prediction. The 2 in this last expression is called the g-factor. This expression for the magnetic moment of the spinning electron can be conveniently expressed as

$$\mu_s = 2\cdot\frac{e}{2mc}\cdot p_s = 2\cdot\frac{eh}{4\pi mc}\cdot s = 2\cdot s\cdot\mu_B ,$$

in which μ_B, called the Bohr magneton, has the value

$$\mu_B = \frac{eh}{4\pi mc}$$

and s, the spin angular momentum of the electron, has the value ½ expressed in units of $\left(\frac{h}{2\pi}\right)$. Wave mechanics offers an adequate explanation of magnetic effects associated with the orbital angular momentum

of electrons, but the explanation of the value observed for the magnetic moment of the spinning electron is explained at the present only by Dirac's relativistic wave mechanics, and this explanation now appears to be too small by 12 parts in ten thousand.[3,4]

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[3] Kusch, P., and Foley, H.M. Phys. Rev. 72, 1256 (1947).

[4] Lamb, W.E., and Retherford, R.C. Phys. Rev. 72, 241 (1947).

By analogy with that of the electron it might appear that the magnetic moment of the proton due to its spin should be $2 \cdot \frac{e}{2Mc} \cdot p_I$, where M is the mass of the proton and p_I the spin angular momentum. If this prediction were correct the proton would have a magnetic moment roughly one two-thousandth that of the electron, since both particles have spin $\frac{1}{2}\left(\frac{h}{2\pi}\right)$ in the ground state. Although this analogy is not borne out by experiment, it is nevertheless convenient to measure nuclear magnetic moments in units of

$$\mu_n = \frac{eh}{4\pi Mc}.$$

The unit μ_n is called the nuclear magneton. Since nuclear magnetic moments differ only in magnitude and possibly sign from the prediction given in

$$\mu_I = \frac{e}{2Mc} \cdot p_I = I \cdot \frac{eh}{4\pi Mc} = I\mu_n,$$

it is common practice to introduce a nuclear g-factor defined by the equation

$$\mu_I = g_I \cdot \frac{e}{2Mc} \cdot p_I = g_I \frac{eh}{4\pi Mc} \cdot I = g_I I \mu_n,$$

where p_I represents $I\left(\frac{h}{2\pi}\right)$. In general g_I is not the same for any

two different types of nuclei.

It is informative to consider the magnetic moment of the deuteron, the simplest compound nucleus. To do this first consider the individual magnetic moments of its constituents, the neutron and the proton. Experiment has revealed that the magnetic moment of the proton is $(2.79268 \pm 0.00006)\mu_n$.[5] Since the spin of the proton is ½, one obtains the value 5.58536 for its nuclear g-factor. The neutron, an uncharged particle by classical theory expected to have zero magnetic moment, nevertheless has been observed to have a spin magnetic moment[6] of $(-1.91280 \pm 0.00009)\mu_n$. Its g-factor is thus seen to be (-)3.8256. In the deuteron the projected spin vectors of the proton and the neutron are parallel and give a value of unity for the nuclear spin. Thus one might suppose that the magnetic moment of the deuteron would be given by the algebraic sum of the moments of the two elementary particles, which would be

$$\mu(D^2) = \mu(P^1) + \mu(n^1) = 0.87988\,\mu_n.$$

However, experiment gives a magnetic moment[7] of $0.85738\,\mu_n$ for the

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[5] Sommer, H., Thomas, H.A. and Hipple, J.A. *Phys. Rev.* *80*, 487 (1950).

[6] Mack, J.E. *Revs. Mod. Phys.* *22*, 64 (1950).

[7] Walchli, H.E. *A Table of Nuclear Moment Data (Preliminary Issue).* 1952, p. 13.

deuteron, a difference of $0.02250\,\mu_n$, which may be considered precise to 0.5%. Thus the last equation is not true. Theoretical explanations for this difference have been offered but the problem can by no means be considered completely solved. Nor has any acceptable theoretical justification yet been proposed for the values obtained by experiment for the magnetic moments of the proton and the neutron nor for any compound nucleus when its spin is other than zero. For most nuclei with higher mass numbers and non-zero spin it is usually impossible to predict even approximately the magnitudes of their magnetic moments.

Present techniques for measuring relative magnetic moments of nuclei commonly give an accuracy of five or six significant figures. This accuracy of measurement is rivaled only by those made with the mass spectrograph, so it is hoped that nuclear magnetic moment determinations may in the future prove to be of as great or greater value in the formulation of nuclear theory as relative mass determinations have been in giving to us our present knowledge of nuclear forces.

(d) Nuclear Quadrupole Moment

The nuclear quadrupole moment is a measure of the extent by which the charge of a nucleus varies from spherical symmetry. The expression for the quadrupole moment may be obtained briefly in the following manner.[8]

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[8] Halliday, D. Introductory Nuclear Physics. New York: John Wiley & Sons, 1950, pp. 59-63.

If a point in space is chosen as an origin and from there a straight line is drawn as a reference axis, the expression for the scalar potential at points along the axis due to a finite number of discrete point charges near the origin can be obtained as a function of R, the distance on the axis from the origin. This scalar potential ϕ can always be expressed by means of the series

$$\phi = c_1 R^{-1} + c_2 R^{-2} + c_3 R^{-3} + c_4 R^{-4} + \cdots,$$

in which c_n is the component of the electrical multipole moment of order 2^{n-1} for the particular configuration, origin and direction. In the above series the quadrupole term is therefore seen to be represented by $\frac{c_3}{R^3}$.

In nuclei the discrete charges are protons, and the nuclear center of mass and the axis of nuclear spin are taken as the origin and the reference axis, respectively.

If the discrete system of charges is changed to a continuous charge distribution of uniform density δ with center of charge somewhere near the origin, one obtains as the expression for the scalar potential the series

$$\phi = R^{-1}\iiint \delta\, dv + R^{-2}\iiint z\,\delta\, dv + R^{-3}\iiint (3z^2 - a^2)\,\delta\, dv + \cdots.$$

In this equation "a" is the radial distance of the volume element, dv, from the center of the charge distribution, and z is the component along the spin axis of the radius vector from the origin to the volume element. The magnitudes of the various order multipole terms give indications of the distribution of charge about the particular origin and axis of reference.

The dipole term will always be zero for any charge distribution possessing mirror symmetry about a plane perpendicular to the axis and passing through the origin. The dipole term is zero for all nuclei.

Nuclei with zero spin have no preferred axis, so the charge distribution is effectively symmetric with the result that such nuclei do not possess quadrupole moments. Wave mechanics can be used to prove that the quadrupole moment is zero also when the nuclear spin has the value one-half.[9] Hence no nucleus with spin less than unity can possess a quadrupole moment other than zero. For other nuclei, a potential function with a negative quadrupole term indicates an oblate spheroidal distribution of charge about the axis of spin, whereas a potential function with positive quadrupole term indicates a prolate spheroidal distribution.

In nuclear theory the quadrupole component is defined as

$$Q = \frac{1}{Ze}\iiint (3z^2 - a^2)\, \delta \, dv.$$

Experimental determinations of Q give values[10] ranging from Q = 7.0 x 10^{-24} cm^2 for $_{71}Lu^{176}$ to (-)1.2 x 10^{-24} cm^2 for $_{51}Sb^{72}$.

It has not yet been possible to demonstrate experimentally any conclusive evidence of octupole or higher order electric moments in nuclei.

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[9] Blatt, J.M., and Weisskopf, V.F. Theoretical Nuclear Physics. New York: John Wiley & Sons, 1952, pp. 26-30.

[10] Mack, J.E. Revs. Mod. Phys. 22, 64 (1950).

II. METHODS OF MEASURING MAGNETIC MOMENTS

(a) Hyperfine Structure

Approximate values of nuclear magnetic moments may be determined from hyperfine structure in atomic spectra by using a method developed by Goudsmit.[11] Since this method involves certain approximations and some correction factors which are themselves inaccurately known, it is perhaps the least accurate method for this purpose. The method remains of value even today, however, for not all nuclear magnetic moments have yet been determined by more precise methods. Such is the case with the isotopes of silver, for which the only values so far obtained are those calculated from hyperfine structure data.

The hyperfine splitting of an energy level with total electronic angular momentum $(Jh/2\pi)$ is given by

$$\delta\nu = AIJ\cos(IJ) = \tfrac{1}{2}A\left\{F(F+1) - J(J+1) - I(I+1)\right\},$$

in which I is the nuclear angular momentum in units of $(h/2\pi)$, F is the resultant of I and J, and the cosine is a "quantum" cosine. The proportionality factor A is equal to the distance between two adjacent hyperfine levels divided by the largest of their F values and is given a positive sign when the larger F value belongs to the higher energy. A is proportional to the nuclear g-factor.

For a single non-penetrating electron it is customary to write

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11. Goudsmit, S. Phys. Rev. 43, 636 (1933).

"a" instead of A and the following relationship holds,

$$a = \frac{R\alpha^2 Z^3}{n^3(l+\frac{1}{2})j(j+1)} \cdot \frac{g_I}{1838}\ cm^{-1}.$$

For a single outer electron in a penetrating orbit, the expression for "a" becomes

$$a = \frac{R\alpha^2 Z_i Z_o^2}{n_o^3(l+\frac{1}{2})j(j+1)} \cdot \frac{g_I}{1838}\ cm^{-1}$$

in which n_o is the principal quantum number, Z_o is the effective nuclear charge when the electron is outside the core electrons, and Z_i the average effective nuclear charge when the electron has penetrated the core.

The ordinary spin doublets for non-s electrons are obtained from the formula

$$\Delta\nu = \frac{R\alpha^2 Z_i^2 Z_o^2}{n_o^3\, l(l+1)}\ cm^{-1},$$

which, combined with the previous equation gives

$$a = \frac{\Delta\nu}{Z_i(l+\frac{1}{2})} \cdot \frac{l(l+1)}{j(j+1)} \cdot \frac{g_I}{1838}\ cm^{-1}.$$

After applying certain relativity corrections shown to be necessary by Breit[12] and by Racah[13] and solving for g_I, one obtains for s-electrons

$$g_I = \frac{3a}{8R\alpha^2}\ \frac{n_o^3}{Z_i Z_o^2} \cdot \frac{1838}{\kappa(\frac{1}{2}, Z_i)}.$$

For non-s electrons, the result is

$$g_I = \frac{a Z_i}{\Delta\nu} \cdot \frac{j(j+1)(l+\frac{1}{2})}{l(l+1)} \cdot \frac{\lambda(l, Z_i)}{\kappa(j, Z_i)} \cdot 1838.$$

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12. Breit, G. *Phys. Rev.* *38*, 463 (1931).

13. Racah, G. *Zeits. f. Physik* 71, 431 (1931).

$\kappa(j,Z_i)$ is the relativity correction by which the equation for the hyperfine structure must be multiplied, and $\lambda(l,Z_i)$ is a similar correction for the multiplet separation. They are determined by the formulas

$$\kappa(j,Z_i) = 4j\,(j+\tfrac{1}{2})(j+1)/(4\rho^2-1)\rho, \qquad \rho^2 = (j+\tfrac{1}{2})^2-(\alpha Z_i)^2,$$

$$\lambda(l,Z_i) = \left[2l(l+1)/(\alpha Z_i)^2\right]\left\{\left[(l+\tfrac{1}{2})^2-(\alpha Z_i)^2\right]^{1/2} - 1 - \left[l^2-(\alpha Z_i)^2\right]^{1/2}\right\}.$$

In many-electron spectra the same electron may be responsible for part or all of the interaction which produces the hyperfine splitting of several different levels. If "a" is calculated for this electron from the various hyperfine structures and these values turn out to be consistent with each other, this agreement of values for "a" is not necessarily sufficient to warrant the assumption that the approximation made in the various formulas above are valid. Only if the values of g_I calculated from different electrons of the same atom agree should one assume that the approximations involved are admissible.

(b) Molecular Beams

In the molecular beam method as developed by Rabi and his coworkers[14] at Columbia University for measuring nuclear magnetic moments, a well collimated beam of neutral molecules is passed through the fields

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[14] Rabi. I.I., Millman, S., Kusch, P., and Zacharias, J.R. Phys. Rev. 55, 526 (1939).

of three magnets, A, C and B, in a highly evacuated space as shown in Figure 1. The source of the molecules is a small oven O which contains the appropriate substance and from which molecules with a modified Maxwellian speed distribution escape through a small slit. These escaping molecules are collimated by additional slits so that all molecules entering the field of magnet A do so within a very small angle to a line parallel to the length of the pole faces. A further defining slit S is located between magnets A and C. At a small distance past the exit from magnet B on the axis defined by the collimating slits is a detector for measuring the intensity of the molecular beam.

The field of magnet A is of the order of 12,000 gauss and is very inhomogeneous (approximately 10^5 gauss/cm.), the magnet field H and the field gradient (dH/dz) in this magnet being in opposite directions. Magnet B is similar to A except that it is arranged to produce a field H in the same direction as, but has its gradient opposite in direction to that of A. Magnet C, which is between A and B, produces a uniform field in the same direction as those of magnets A and B. A hairpin loop to which can be applied a very weak oscillating radio-frequency field at right angles to the magnetic field is installed between the pole faces of magnet C.

In molecular beam experiments one employs molecules in a $^1\Sigma$ state in which the resultant electronic angular momentum is zero to the first order. It has been calculated that the interactions between the nuclear magnetic moments in a molecule in a $^1\Sigma$ state and between those and the magnetic moment associated with molecular rotation involve

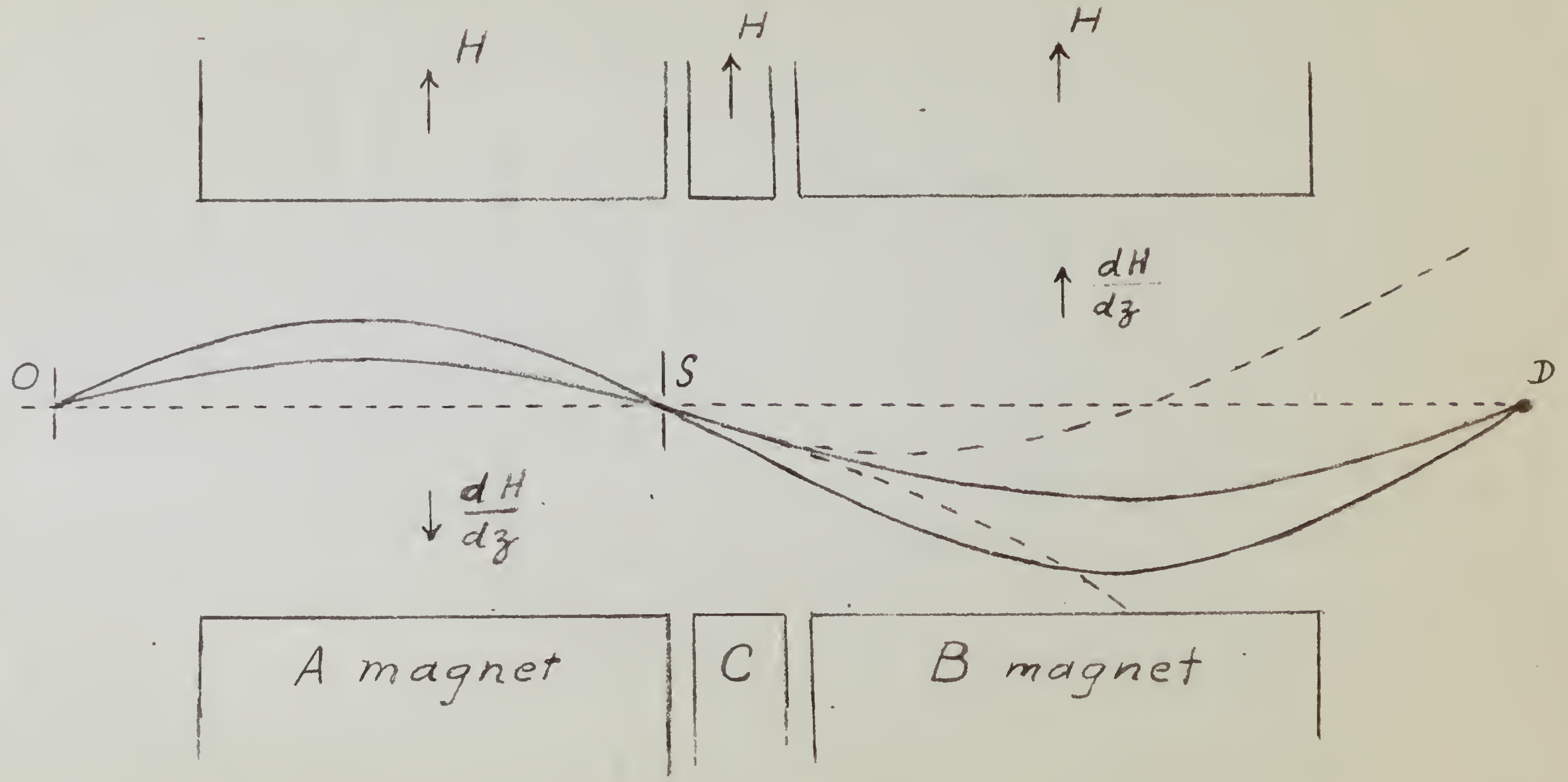

FIGURE 1.
THE PATHS OF FOUR MOLECULES IN A MOLECULAR-BEAM MAGNETIC RESONANCE APPARATUS

magnetic fields of the order of 100 gauss or less.[14] External magnetic fields of several thousand gauss will decouple the nuclear spins from each other and also from the molecular rotation to the extent that the nuclear spins may be regarded as free.

A nucleus of spin I when in a magnetic field may assume one of only 2I+1 different directions with respect to the direction of the magnetic field.* In an inhomogeneous magnetic field of the type shown for magnets A and B a nucleus with magnetic moment experiences a resultant force $\mu_z \frac{dH}{dz}$, where μ_z is the component of μ in the direction of the field H.

Consider now a molecule which has entered the field of magnet A with just the proper velocity, spin orientation and deviation from the dotted axis shown in Figure 1 to be deflected so as to pass through slit S into the homogeneous field of magnet C. If the nuclear spin undergoes no change in spatial quantization while the molecule is passing through the field of C, the arrangement is such that the molecule will be deflected back toward the axis just sufficiently to impinge upon the detector D after emerging from the field of the third magnet.

After operating conditions have been properly adjusted and the beam of molecules impinges on the detector D, a radio frequency oscillation is applied to the hairpin loop in the field of magnet C.

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*Some of the basic concepts introduced briefly here will be treated in considerable detail in the next section.

Now, the energy difference ΔE between adjacent levels resulting from the nuclear spin axis changing its projection in the direction of the magnetic field by $\Delta M_I = \pm 1$ is

$$\Delta E = g_I \mu_n H.$$

It therefore appears plausible that the proper application of a radio-frequency field of frequency given by $h\nu = \Delta E$ might produce transitions between adjacent levels. Indeed, transitions involving reorientation of the nuclear spin vector with respect to the magnetic field involving either emission or absorption are induced when the frequency of the r-f oscillations applied to the hairpin loop has the resonance value

$$\nu = \frac{\Delta E}{h} = \frac{g_I \mu_n H}{h}.$$

Thus when the proper field strength H exists for the r-f oscillations to be of the frequency required for resonance, transitions involving reorientation of the direction of nuclear spin axes will occur. Molecules whose nuclei undergo such reorientation of spin axis while passing through the field of magnet C will no longer be deflected by magnet B so as to impinge upon the detector but will be deflected so as to miss it as shown by the dotted trajectories in Figure 1. Thus, when the resonance condition is realized there is a decided decrease in detector "current". The nuclear g-factor of the isotope being studied may then be obtained from the relations

$$\Delta E = h\nu = g_I \mu_n H, \qquad \text{and}$$

$$g_I = \frac{h\nu}{\mu_n H}.$$

From the above relations it will be noted that the frequency of radiation required to produce transitions in the field of O is independent of the particular orientation of the nuclear spin axis. For most nuclides the resonance frequency is of the order of several megacycles per second for fields of some 2,000 to 12,000 gauss.

The Rabi method was the first to give values of nuclear magnetic moments to four significant figures. The limitation upon accuracy in this method is the requirement that the strength of the magnetic field be determined. Other disadvantages of the molecular beam method are the requirements for a high vacuum throughout the magnet system and for a fairly high temperature in the oven. But it also has certain advantages, among them being the fact that very minute quantities of an isotope are required for measurements.

This method has largely been replaced by the more accurate nuclear resonance spectrometers, but its usefulness has by no means disappeared.

(c) Work on Liquids and Solids

The next significant advance in the technique of measuring nuclear magnetic moments came in 1946 when Purcell, Pound and Torrey at Harvard[15] and Bloch, Hansen and Packard at Stanford[16] simultane-

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[15] Purcell, E.M., Torrey, H.C., and Pound, R.V. *Phys. Rev.* *69*, 37 (1946).

[16] Bloch, F., Hansen, W.W., and Packard, Martin. *Phys. Rev.* *69*, 127 (1946).

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cusly and independently developed new resonance techniques requiring equipment appreciably more simple than that used in molecular beam methods. A distinct advantage of these new methods lies in the fact that it frequently is unnecessary to alter the physical or chemical form of the sample, thus avoiding many of the experimental difficulties of the molecular beam method. In addition to their use in nuclear studies these new methods provide a means for investigating the establishment of the thermal equilibrium essential to magnetic methods for attaining very low temperatures and for obtaining information concerning crystal structure, phase transitions in solids, and hindered internal motions in solids.

The basic semi-classical concept which most readily describes the essential phenomena involved in resonance experiments will now be discussed in detail.

As previously noted, the nuclear magnetic moment may be expressed as

$$\mu_I = g_I \cdot \frac{e}{2Mc} \cdot p_I .$$

Results from wave mechanics indicate that the value of $|\underline{p}_I|$ is $[I(I+1)]^{\frac{1}{2}}\left(\frac{h}{2\pi}\right)$ but that the maximum projected value which $|\underline{p}_I|$ may assume in the direction of an applied external magnetic field is

$$(p_H)_{max} = I\left(\frac{h}{2\pi}\right).$$

The permitted values of p_I, the projected magnitude of $\underline{p}_I$ upon the direction of the field, are

$$p_I = M_I\left(\frac{h}{2\pi}\right),$$

where $M_I = I, I-1, I-2, \cdots, -I+1, -I$.

If a magnet of dipole moment $\underline{\mu}$ is placed in a magnetic field $\underline{H}$,

there is a torque $\underline{L}$ exerted on the dipole given by the expression[17]

$$\underline{L} = \underline{\mu} \times \underline{H}.$$

Since the rate of change of angular momentum, $\underline{p}$, of a system is equal to the applied torque, there results

$$\frac{d\underline{p}}{dt} = \underline{L},$$

or

$$\frac{d\underline{p}}{dt} = \underline{\mu} \times \underline{H}.$$

The vector expression for the nuclear magnetic moment is

$$\underline{\mu}_I = g_I \cdot \frac{e}{2Mc} \cdot \underline{p}_I,$$

so for a nuclear dipole in a magnetic field one may write

$$\frac{d\underline{p}_I}{dt} = -g_I \cdot \frac{e}{2Mc} \cdot \underline{H} \times \underline{p}_I.$$

From classical mechanics we also have the vector relationship

$$\frac{d\underline{p}}{dt} = \underline{\omega}_0 \times \underline{p},$$

so it may be concluded that if a nucleus of magnetic moment vector $\underline{\mu}_I = g_I \cdot \frac{e}{2Mc} \cdot \underline{p}_I$ is placed in a magnetic field, the magnetic moment vector of the nucleus will precess about the direction of $\underline{H}$ with the angular frequency

$$\omega_0 = -g_I \cdot \frac{e}{2Mc} \cdot \underline{H}.$$

This frequency of precession is called the Larmor precession frequency. From the last equation it is noted that $\underline{\omega}_0$ is not dependent on the angle between $\underline{\mu}_I$ and $\underline{H}$.

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[17] Pake, G.E. Am. Jour. Phys. 18, 438 (1950).

The potential energy U of a magnetic dipole $\underline{\mu}$ in a field $\underline{H}$ is $U = |\underline{\mu}||\underline{H}|(1-\cos\theta)$, where θ is the angle between $\underline{\mu}$ and $\underline{H}$. This expression gives a maximum for U of $2|\underline{\mu}||\underline{H}|$ when $\underline{\mu}$ and $\underline{H}$ are antiparallel. Since the nuclear angular momentum vector may assume 2I+1 orientations with respect to $\underline{H}$, with M_I = I, I-1, ..., -I+1, -I, there will be in all 2I energy levels due to nuclear spin orientations, each differing in energy by

$$\frac{2(\mu_H)_{max} H}{2I} \quad \text{or} \quad \frac{(\mu_H)_{max} H}{I} \text{ ergs.}$$

The selection rule for transitions between the 2I+1 possible orientations of the magnetic moment vector is

$$\Delta M_I = \pm 1.$$

This requires that the nucleus emit or absorb

$$\Delta E = h\nu = \frac{(\mu_H)_{max} H}{I} \text{ ergs.}$$

in event of change of orientation of spin vector. The frequency obtained from this expression is seen to be

$$\nu = \frac{(\mu_H)_{max} H}{Ih}.$$

But

$$\frac{(\mu_H)_{max} H}{Ih} = g_I \cdot \frac{e}{2Mc} \cdot I \frac{h}{2\pi} \cdot \frac{1}{Ih},$$

or

$$\nu = g_I \cdot \frac{e}{4\pi Mc} \cdot H$$

Using $\omega = 2\pi\nu$ one obtains

$$\omega = g_I \cdot \frac{e}{2Mc} \cdot H,$$

which is the absolute value previously found for the Larmor angular precession frequency. In other words, an oscillating electromagnetic field of frequency

$$\nu = g_I \cdot \frac{e}{4\pi Mc} \cdot H$$

applied to a nucleus in the external magnetic field $\underline{H}$ might be capable of inducing transitions between adjacent nuclear spin orientation states. In the following four sections there will be discussed the four most recent methods developed for applying the above principle.

All of the four methods depend upon the Maxwell-Boltzmann relation that, in any system in thermal equilibrium, the ratio of the populations of states with different energies, say E_1 and E_2, is

$$\frac{N(E_1)}{N(E_2)} = \frac{e^{-E_1/kT}}{e^{-E_2/kT}}.$$

If this relation is applied to the case of protons in a magnetic field H, the ratio of the populations of the two proton states $M_I = +\frac{1}{2}$ and $M_I = -\frac{1}{2}$ is seen to be

$$\frac{N(+\frac{1}{2})}{N(-\frac{1}{2})} = \frac{e^{-E_1/kT}}{e^{-E_2/kT}} = e^{(E_2-E_1)/kT} = e^{\Delta E/kT},$$

in which $(E_2 - E_1) = \Delta E = g_I \mu_n H.$

For temperature T = 300°K and magnetic field strengths available in laboratories $\frac{\Delta E}{kT}$ is small, so the last equation may be written

$$\frac{N(+\frac{1}{2})}{N(-\frac{1}{2})} \cong 1 + g_I \mu_n \frac{H}{kT}.$$

For a field of 10,000 gauss, $g_I = 5.58$ for protons, and $kT = 4\times10^{-14}$ erg the last expression gives

$$\frac{N(+\frac{1}{2})}{N(-\frac{1}{2})} \cong 1 + 7\times10^{-6}.$$

This indicates that for each million protons in the higher energy state there are one million and seven protons in the state

with lower energy. Clearly this relationship may be extended to nuclei with spin I possibly other than $\frac{1}{2}$. The results of the above example will be referred to in some of the forthcoming discussions.

Another important factor in the methods to be discussed is the spin -lattice relaxation time T_1 for the nucleus of interest in a given liquid or solid. Thermodynamically, a relaxation process in the case of nuclei is any method of energy exchange between the system of nuclear spins and the lattice. Thus if a sample is placed suddenly into a magnetic field it is important for the experimenter to know how long a period of time on the average will elapse before the equilibrium number of nuclei will be found in the upper and lower energy states. T_1 is employed as a measure of this time and is defined as the length of time required for all except the fraction (1/e) of the equilibrium excess number of nuclei to reach the lower energy state. Relaxation mechanisms in fluids have been investigated by Purcell and his group[18] at Harvard who have determined that Brownian motions at the Larmor frequency are responsible for the processes. In liquids T_1 ordinarily decreases with increase of viscosity. In some cases, however, T_1 decreases with increasing viscosity until a minimum value is reached and after that it increases with increase in viscosity.

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[18] Bloembergen, N., Purcell, E.M., and Pound, R.V. Phys. Rev. 73, 679 (1948).

The addition of paramagnetic ions to a sample frequently will give a marked decrease in T_1 by increasing the local perturbing effects upon nuclei of the sample.

(1) Method of Purcell and Pound

In the method developed by Purcell, Pound, and Torrey,[18] nuclear absorption unbalances an r-f bridge, giving rise to a signal in a receiver used as a detector. The sensitivity of this method is very good, being as high perhaps as any of the other three methods discussed in this section. Purcell has demonstrated that nuclear resonance with this method can be observed in a sample containing only 10^{19} atoms.

In this method a signal generator is employed as the r-f signal source for the bridge. An essential feature of the bridge is the reduction effected in the relative magnitude of output fluctuations arising from amplitude fluctuations in the signal supplied by the r-f signal generator. This is done by obtaining a voltage node in the bridge output to the amplifier when the bridge is placed in balance with the system not adjusted for nuclear resonance. One method to effect this voltage node has been to place an extra half-wave-length of cable in one arm of the bridge. Another important feature of this method is the reduction in the r-f level at the input to the amplifier to permit considerable r-f amplification before detection.

One arm of the bridge contains a tuned circuit having in the gap between the pole faces of an electric magnet a coil into which a

sample to be studied can be inserted. In the other arm of the bridge is a dummy circuit similar in all respects to the other but not having its coil located in the magnetic field. Nuclear resonance absorption in the sample causes a change in the balance of the r-f bridge which is detected by means of the receiver. When liquids of high dielectric constants are being studied a dummy sample is sometimes placed in the dummy coil to balance as nearly as possible the appreciable change in stray capacitance resulting from the sample being placed in the coil of the other arm.

The strong magnetic field H_0 is modulated sinusoidally at 30 cycles per second by auxiliary coils mounted on the pole pieces. This modulation is parallel to the main field with peak strength never more than 15 gauss. Near resonance the low frequency modulation of the magnetic field causes the Larmor frequency of nuclear precession to vary in and out of resonance with the result that resonance absorption occurs in the arm of the bridge containing the sample and the bridge is thrown out of balance. The signal so produced is amplified and suitably detected.

An improvement in the above technique has been the elimination of the half-wave-length cable, which is several meters long and is frequently the cause for considerable troublesome instability.

With properly adjusted equipment operating under suitable conditions the sensitivity of this method depends on the width of the observed resonance. The natural width of the resonance signal depends on the substance used and in some liquids is less than a thousandth of a gauss. This may cause the homogeneity of the magnetic field to be a limiting factor in the sensitivity.

The r-f bridge nuclear resonance spectrometer is well suited for the observation of shapes and widths of absorption lines in various substances. Four such resonance absorption lines for protons in different substances which were obtained by Purcell using the bridge method are shown in Figure 2.[19] The line displayed in Figure 2(a) was obtained from protons in water. This absorption occurred within an interval of only 0.2 gauss in a field of 6,940 gauss. Other substances containing protons display an absorption line at the same field strength if the radio frequency is the same, but the line width varies greatly with different substances. The proton resonance in ice, shown in Figure 2(b), is actually 20 to 50 times as wide as the line obtained in water. In other crystals the variation in line width is even more striking and a pronounced difference in line shapes is observed as one may notice by comparing the various absorption lines shown in this Figure.

The variations in line widths and line shapes find no explanation from nuclear properties but are associated with the perturbing magnetic fields which have their origins in the immediate surroundings of a given hydrogen atom and depend on both the location and motion of its immediate neighbors. It perhaps seems strange, but the sharpest resonance lines are obtained from liquids, whose molecules compared to those of solids are in a greater state of random motion. This random thermal motion of the molecules occurs at a

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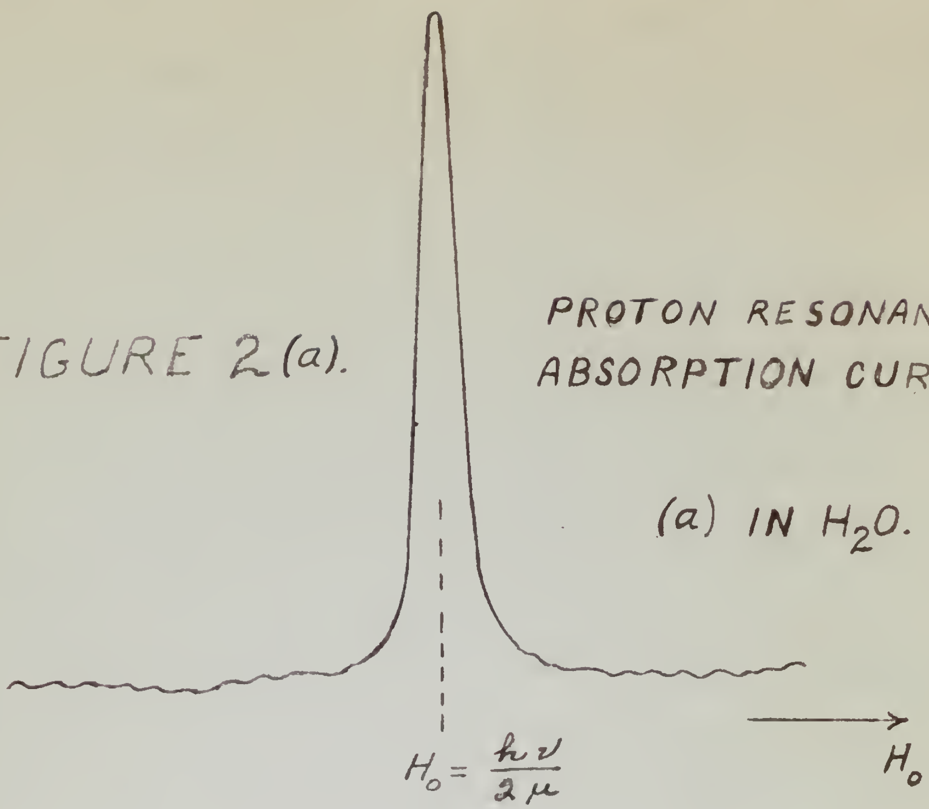
[19] Purcell, E.M. Science 107, 433 (1948).

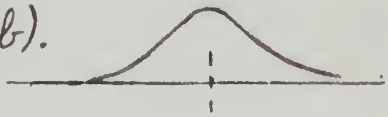
PROTON RESONANCE ABSORPTION CURVE

FIGURE 2(a).



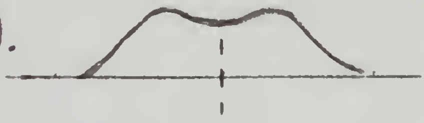
(a) IN H_2O.

FIGURE 2(b).



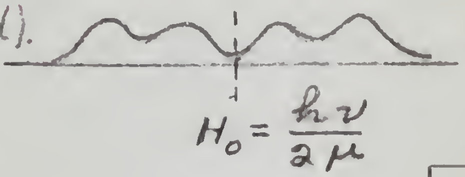
(b). IN ICE.

FIGURE 2(c).



(c). IN POWDERED GYPSUM.

FIGURE 2(d).



(d). IN SINGLE GYPSUM CRYSTAL.

Curves Not to Same Scale. (After Purcell[19])

frequency higher than that of the proton's nuclear precession. It has been shown statistically that the very rapidity of this thermal motion greatly reduces the perturbing effect from neighboring atoms, so that local perturbing effects on the applied magnetic field are much less than in the case of solid substances.[18]

In the case of a single crystal of gypsum referred to in Figure 2(d), protons in different parts of the molecule have different, more nearly constant local perturbing fields which roughly are the same on corresponding protons in different molecules. This results in a broad line with four resonance peaks corresponding to four different values of field strength observed.

In the case of powdered gypsum a greater degree of randomness of orientation of molecules within the sample occurs. The statistical effect of this (imperfect) randomness is to cause the absorption curve to exhibit now two peaks and to contract somewhat in width, as shown in Figure 2(c).

In ice each proton is surrounded by several equally near magnetic neighbors. Also, a certain amount of local motion persists in ice well below the freezing point. This combination of more or less equidistant spacing of magnetic neighbors and of local molecular motion causes a further contraction of line width and a line of only one peak; however, this line is much lower and broader than that of protons in a water sample.

The data obtained from nuclear resonance absorption studies such as the above have yielded some information of the solid state not previously obtainable by other methods. For example, with the aid of certain advanced theory using data obtained by these methods

it has been possible to determine not only the directions of the H-H lines with respect to the axes of the gypsum crystal, but also, with a precision of about 1 per cent, the distance between the two protons in the HOH molecule.[19] X-ray analysis does not yield this information.

(2) Method of Bloch

In the Bloch method for detection of nuclear magnetic resonance there is employed a nuclear induction transformer in which the establishment of nuclear resonance causes a change in coupling between the primary and secondary of the transformer and thus a change in the output voltage. This method of nuclear induction is based on the following principle.[20,21]

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20. Bloch, F. Phys. Rev. 70, 460 (1946).

21. Halliday, D. Introductory Nuclear Physics. New York: John Wiley & Sons, 1950, pp. 518-520.

If a sample of water or paraffin, say, is placed in a strong magnetic field H_0 there will be a small resultant magnetic moment due to the nuclear moments of the protons. The gross external effect results entirely from those few protons, about 7 in each 2,000,000 at a field strength of 10,000 gauss, that represent the difference in population between the two states available to the proton. If the strong magnetic field H_0 is in the z-direction and a weak r-f oscillating field of angular frequency ω is applied at right angles

to H_o in the direction of the x-axis with instantaneous magnitude given by

$$H_x = \hat{H} \cos \omega t,$$

then the combined fields H_o and H_x will produce magnetic polarization in the sample. This polarization may be represented by the vector $\underline{M}$, the magnetic moment per unit volume. Bloch has shown [20] with the aid of certain simplifying assumptions that for a fixed H_o and a fixed ω, (1) the magnitude of $\underline{M}$ is constant, (2) $\underline{M}$ rotates about H_o at the angular rate ω, and (3) $\underline{M}$ makes a fixed angle θ with the H_o axis. The three components of $\underline{M}$ are

$$M_x = M \sin\theta \cos \omega t,$$
$$M_y = M \sin\theta \sin \omega t, \quad \text{and}$$
$$M_z = M \cos\theta,$$

where $|\underline{M}| = M$ and the angle θ is given by the relation

$$\tan\theta = \frac{\hat{H}}{2(H_o - H_{or})},$$

H_{or} being the value of H_o at nuclear resonance. If $H_o \gg H_{or}$, since $\hat{H}$ is not large, θ will also be fairly small. Under the condition of nuclear magnetic resonance, however, $H_o = H_{or}$ and θ becomes 90^o. Consequently at resonance the components of M become

$$M_x = M \cos \omega t,$$
$$M_y = M \sin \omega t, \quad \text{and}$$
$$M_z = 0.$$

From the last equation it is seen that $M_z = M\cos\theta$, which is the

time independent part of M, vanishes at resonance. Since M_x and M_y do not vanish, however, M is perpendicular to the direction of H_o and rotates about it at a frequency which is equal to that of the Larmor precession. This state with $\theta = 90^\circ$ corresponds to equal populations being in each of the two energy levels so that no resultant steady moment exists. In the derivation of this Bloch assumed that the nuclei are entirely free with no spin-lattice interaction. Since spin-lattice interaction is not zero in practice, this equalization with $M_z = 0$ at resonance is not a completely accurate picture of the situation existing at that time.

At the condition of nuclear magnetic resonance the component of M perpendicular to the direction of H_o increases rather suddenly in magnitude. This component of M rotates with the angular frequency ω. As a result, a variable flux links a pickup coil which has its axis in the y-direction. The resultant e.m.f. induced in the pickup coil is passed to a receiver and measured.

The operation of this type of nuclear resonance spectrometer has been summarized in the following manner by the developers of this technique.[22]

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[22] Bloch, F., Hanson, W.W., and Packard, M. Phys. Rev. 70, 479 (1946).

"A spherical sample is immersed in a field $H_{dc} + H_{ac} \cos 377t$ produced by a magnet. The nuclear moments oriented by this field are caused to precess by a driving field H_x. The precessing moments

induce voltages in a receiver coil. These voltages, which vary in amplitude at a 60 cycles per second rate because of the variations in field H_0, are amplified along with stronger constant amplitude signals. The leakage and the varying voltages due to precessing nuclei are mixed in the detector, the output of which then contains pulsating unidirectional current, the steady component due to leakage and the variations corresponding to the desired signal. The d.c. is removed by blocking condensers in the amplifier which increases the signal voltages to a magnitude suitable for operation of the cathode ray tube."

One definitely advantageous feature of this method is that it affords the opportunity to determine the sign of the magnetic moment of a nucleus. This method is suited also for the study of line widths and shapes.

(3) Super-regenerative Method

In the super-regenerative method of detecting nuclear resonances a super-regenerative oscillator is used both to establish nuclear resonance and to detect the induced voltage from it. Several variations in experimental arrangements have been adopted by different investigators using this method. Roberts[23] has described one such arrangement. The apparatus with which the writer obtained his

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[23] Roberts, A. Rev. Sci. Inst. 18, 845 (1947).

measurements was similar in basic principle to that described by Roberts but differed in several details. The principle of operation to be described for the apparatus of this section will apply equally well to either arrangement.

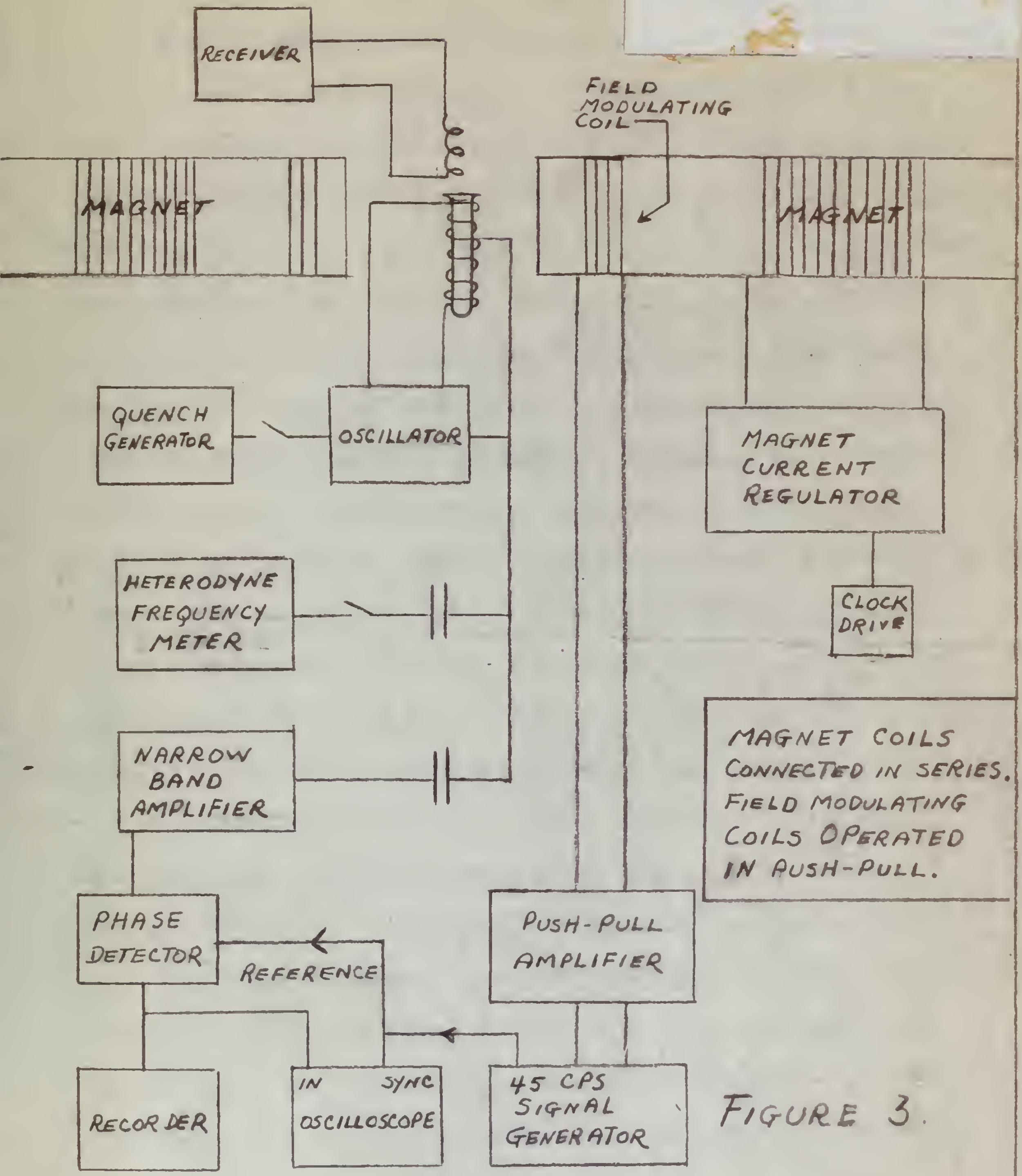
The general layout of the apparatus used by the writer is exhibited schematically in the diagram of Figure 3. The oscillator shown in the diagram with connections to a coil in which a test tube has been inserted is actually placed in its entirety between the pole faces of the large magnet. This oscillator is of the super-regenerative type and from that fact this method derives its name.

In the super-regenerative method the triode of the super-regenerative oscillator produces self-sustained oscillations. For this, adequate feed-back from plate to grid is required to supply all the losses in the circuit. However, instead of allowing continued self-sustained oscillation, an externally generated modulating signal of appropriate strength called the quench voltage is applied either to the grid or to the plate of the triode. This sinusoidal quench voltage causes the overall gain of the triode to be varied in such a way that the self-sustained oscillation of the triode starts and stops once during each quench cycle.

Detection of nuclear magnetic resonance is accomplished in the following manner. When the quench voltage varies from a value which prevents oscillation to one which permits it to begin, the envelope of the current in the tank circuit begins to increase approximately exponentially and, if not inhibited, would reach a saturation value at which feed-back energy would just equal losses. The maximum

BLOCK DIAGRAM

OF APPARATUS



MAGNET COILS CONNECTED IN SERIES. FIELD MODULATING COILS OPERATED IN PUSH-PULL.

FIGURE 3.

amplitude of oscillation reached and the integrated energy of the oscillation pulse are functions of the effective Q of the resonant circuit, the feed-back gain, and the quench amplitude and frequency.

When the oscillator is placed between the pole faces of a magnet and a sample containing the desired type of nucleus, say protons, is placed in the inductance coil of the tuned circuit, the nuclear spin axes of the protons will precess in the direction of the magnetic field at the Larmor frequency. When the magnetic field strength is of such value that the Larmor and the oscillator frequencies are the same, the r-f oscillation at the Larmor frequency will induce emission or absorption of radiation by the protons involving reorientation of their nuclear spin axes in the magnetic field. Since at thermal equilibrium in, say, a field of 10,000 gauss there are in each 2,000,000 hydrogen atoms some 7 more protons in the lower energy than in the higher energy nuclear spin state, there will be a net absorption of r-f radiation by the protons, the effective resistance of the tuned circuit will be increased and there will be a decrease in the rate at which oscillation builds up. Thus the integrated pulse will be less under conditions of resonance, a feature of the super-regenerative method which may be detected by appropriate methods to allow observation of the nuclear resonance.

The quench frequencies employed by the writer in his measurements were of the order of 2 to 10 kilocycles making the quench periods 100 to 500 microseconds. The thermal relaxation time for protons in water at room temperature has been measured[24] and found

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24. Hahn, E.L. Phys. Rev. 76, 145 (1949)

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to be 2.33 ± 0.07 seconds. Thus, in the case of the water sample, during that period of the quench cycle when the tuned circuit is permitted to oscillate there will be a very few more protons raised to the higher energy level than will be lowered to the level of lesser energy. During the quenched period there will be a tendency for thermal equilibrium to be re-established among nuclei, so when oscillation is again initiated more protons will again be elevated than lowered in energy state. This process gives a net absorption of energy from the super-regenerative oscillator for each cycle of the quench oscillations. It is this net absorption of energy which results in the Q of the circuit being lowered and the detection of resonance by the super-regenerative method being made possible.

Another feature of this method which should be noted is that the buildup of oscillation in the super-regenerative oscillator is exponential or faster and, if it starts from residual noise voltage in the tank circuit, may possibly cover a significant fraction or all of the quench cycle period without reaching saturation. If, however, when the quench cycle commences there is in the tuned circuit an external signal of the same frequency, the oscillation will build up from this external signal and either will reach a larger amplitude within the length of time permitted by the quench cycle, or else will reach saturation sooner. Although the external signal does not affect the rate of buildup of oscillation, the integrated pulse energy will nevertheless be greater. This can possibly be of

importance because of the following effect.

At the condition of resonance the nuclear spin axes of the sample precess about the direction of the magnetic field with the same frequency as the r-f oscillation of the tuned circuit. This precession continues during the period when the triode oscillation is quenched. When the quench voltage again reaches a value which allows oscillation to be resumed, it is possible that an e.m.f. may be induced in the oscillator coil as a result of persisting precession of the vector $\underline{M}$ as described in the Bloch method. If so induced this e.m.f. will alternate at the Larmor frequency which, at the resonance condition, would provide an external signal of the correct frequency to cause oscillation to start building up immediately in the tuned circuit. Being started immediately in this manner causes the oscillation to reach a larger amplitude or perhaps to reach saturation sooner than would be the case if the oscillation were built up from random noise. Thus in the event that the oscillation is built up from an induced e.m.f. due to nuclear precession, the integrated energy pulse of the oscillation will differ from that of non-resonance.

In either case, whether the Q of the circuit is changed at resonance or whether the precessing nuclei act like a signal generator in the tuned circuit of the oscillator, the integrated pulse of the r-f oscillation will be different from that of the non-resonance condition and thus may be detected and nuclear resonance observed. It is entirely possible that both effects may occur simultaneously in varying proportions under usual operating conditions.

The spectrum of a super-regenerative nuclear magnetic resonance

detector is not a single line spectrum but consists of a central resonance signal corresponding to the fundamental resonance frequency ν_0 and usually one or more pairs of sidebands. These sidebands occur at frequencies ν_s, given by

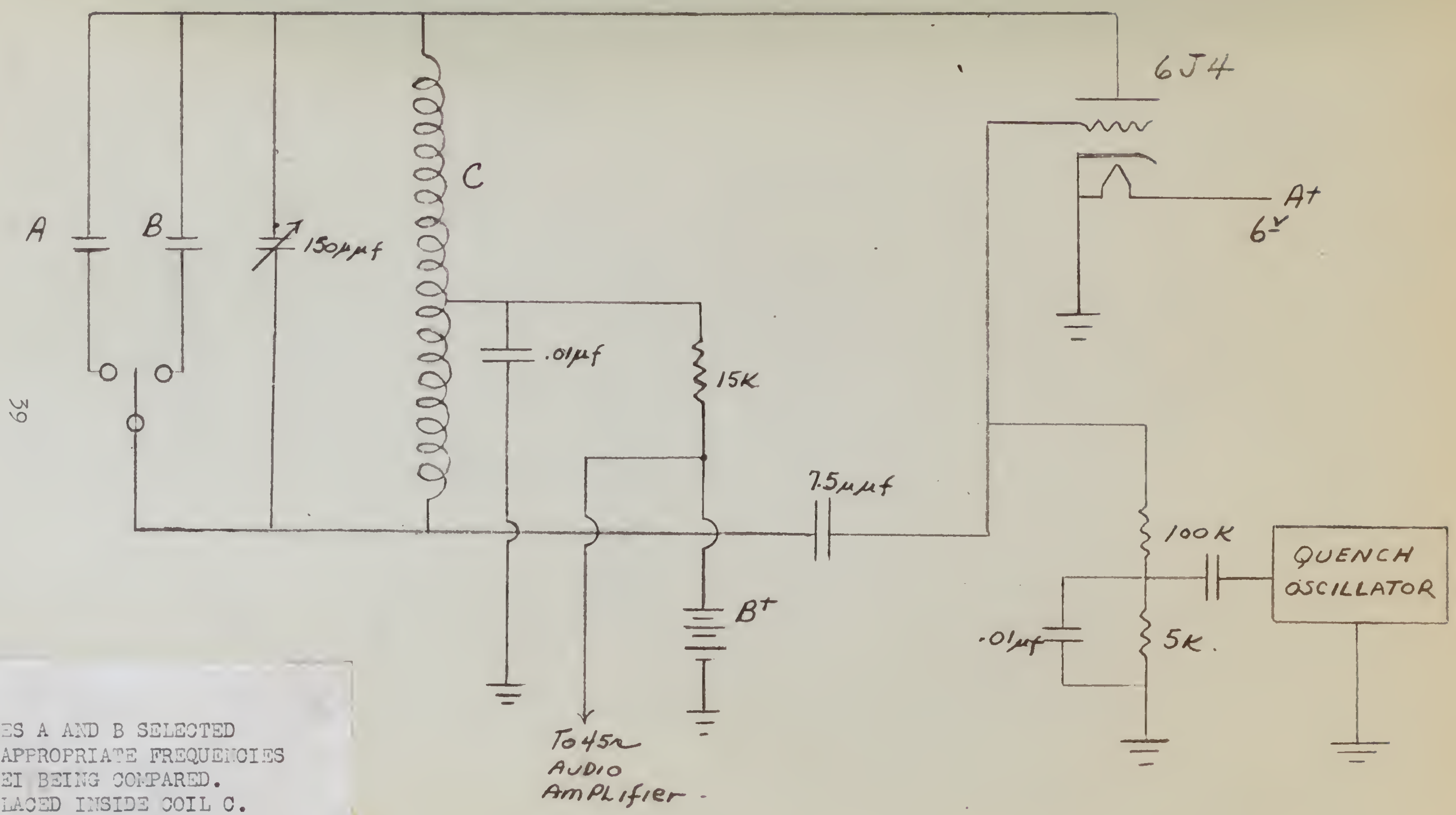
$$\nu_s = \nu_0 \pm n\nu_{qu},$$

in which ν_{qu} is the frequency of the a.c. quench voltage and assumes the values of successive small integers. Resonance absorption by the sample occurs and under proper test conditions may be detected as the magnetic field passes through that strength necessary to produce Larmor precession of the nuclei at the frequency ν_s for each of this limited number of sidebands.

The circuit of the super-regenerative oscillator used by the writer is shown in the diagram of Figure 4. In this circuit the quench frequency voltage is applied to the grid of the oscillator triode. Certain other investigators have applied the quench voltage to the plate. This last arrangement of the quench voltage at one time was used with the apparatus employed in this work but was abandoned because of the high noise to signal ratio resulting from this arrangement. An oscillator circuit of this latter type with quench voltage applied to the plate of the triode as used by Roberts[23] is shown in Figure 5.

(4) Heterodyne Method

In the heterodyne method a weakly oscillating detector is modulated by the nuclear absorption. Such a weakly oscillating detector



CAPACITIES A AND B SELECTED TO GIVE APPROPRIATE FREQUENCIES FOR NUCLEI BEING COMPARED. SAMPLE PLACED INSIDE COIL C.

Pole Face Super-Regenerative Oscillator

FIGURE 4

THE SUPER-REGENERATIVE RESONANCE DETECTOR (AFTER ROBERTS)[23]

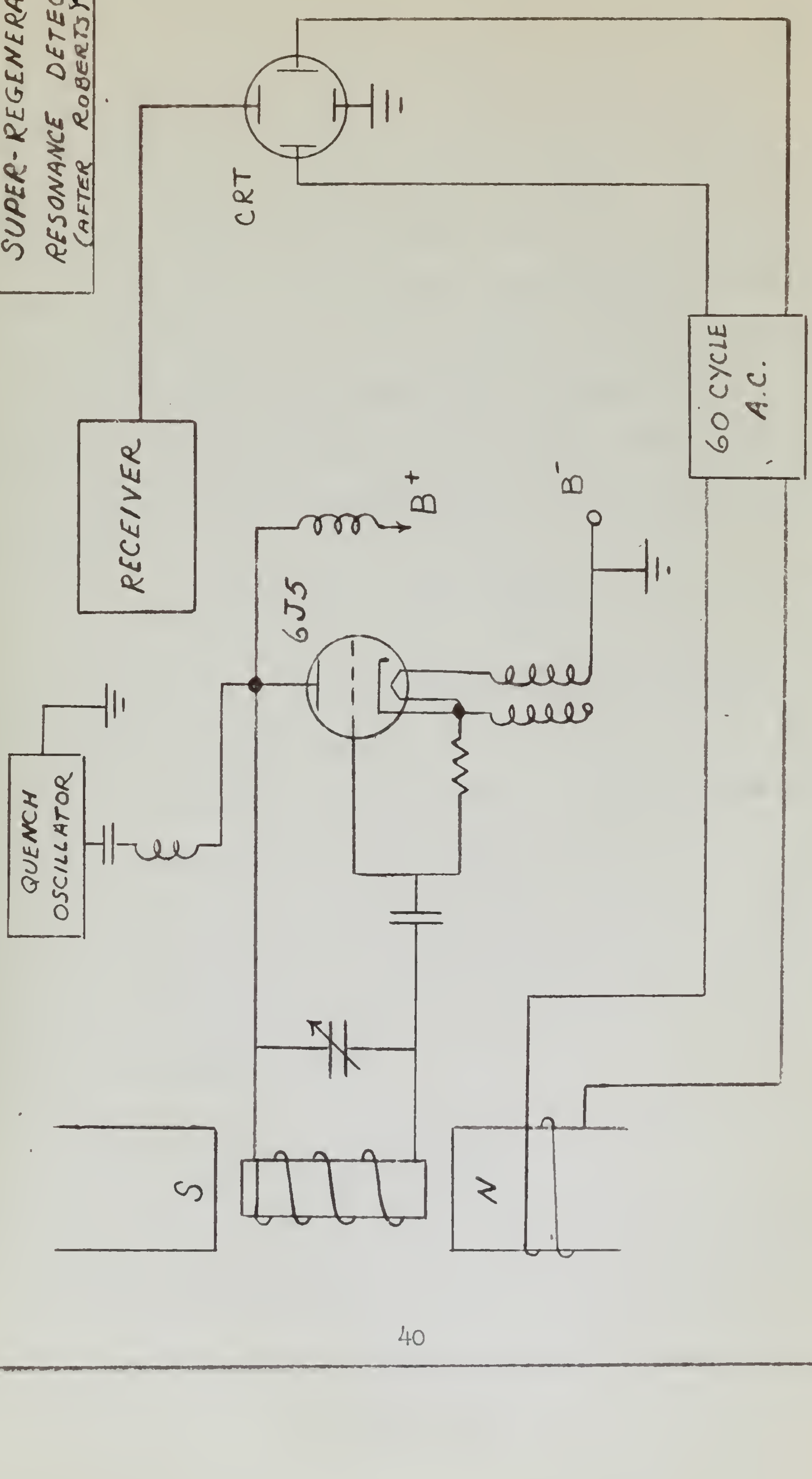


FIGURE 5.

is called an autodyne; in this case the signal from the autodyne beats against an incoming c-w signal to produce a heterodyne signal. A frequently difficult but required condition is that the oscillation of the autodyne be extremely weak, for in this method the strength of the resonance signal is found to diminish as that of the autodyne signal increases, apparently because of saturation of the nuclear absorption. This saturation evidently results from the fact that the total power which can be absorbed by a sample small enough to allow suitable homogeneity of the magnetic field is fairly minute. For N atoms with spin I at temperature T whose Larmor frequency is ν and relaxation time T_1, the power which can be absorbed is

$$P = \frac{2I}{I+1} \cdot \frac{N}{T_1} \cdot \frac{(h\nu)^2}{kT}.$$

For a sample of 1 c.c. of water at 300°K with ν_0 at 30 mc/sec, P will be approximately 10^{-9} watts.[25]

The super-regenerative circuit shown in Figure 5 can be adapted for use as an autodyne detector if the quench voltage supply is removed and the plate voltage properly adjusted. The circuit shown in this figure would probably not be as sensitive as a super-regenerative oscillator.[25] However, improved and more sensitive forms of this method have been devised.[25]

Nuclear magnetic resonance may be observed in either of two

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25. Pound, R.V. Phys. Rev. 72, 527 (1947).

ways when using the autodyne detector. If the output is viewed by an a-m receiver the absorption curve for a liquid sample appears much the same as that of the absorption line in Figure 2 (a). If f-m detection is used, the signal has the form of an anomalous dispersion curve. This f-m effect results from the oscillator being slightly detuned by nuclear resonance effects.

III. EXPERIMENTAL ARRANGEMENTS

(a) Apparatus

The experimental arrangement used in conducting the tests described in this report is indicated in the block diagram of Figure 3. The magnet used to produce the strong magnetic field was a double yoke type with high voltage, low current windings and an adjustable air gap. The gap width used in these experiments was approximately 3/4". Pole pieces of $3\frac{1}{2}$" diameter were used in early work, but all measurements included in this report were made with 6" pole pieces because of the better magnetic field homogeneity obtainable with the larger diameter pole faces. The magnet current was supplied by an electronically controlled current regulator with a clockdrive mechanism operating a potentiometer in the control circuit. This current regulator was capable of increasing or decreasing the magnetic field at a rate of about 2 gauss per minute over a field strength range of some 2,000 to 10,000 gauss. Except under unfavorable conditions seeming usually to stem at irregular hours from rapid uneven variations in the 110 volt 60 cycle a.c. power supplied to the laboratory, the current regulator was capable of maintaining the magnet field current constant to about one part in thirty thousand.

The strong magnet field H_o was modulated by a 45 cycles per second current applied to small field modulating coils called "sweep coils," one being clamped on each pole piece. The 45 cycle current for these windings was supplied by a signal from an audio frequency signal generator amplified by a push-pull amplifier. This small

alternating modulation H_m to the strong field H_0 caused the resonance signal to be repeated periodically at any time when the resonance frequency happened to be within the limits corresponding to the field range $H_0 \pm H_m$. The resonance signal was observed usually on an oscilloscope with its sweep synchronized to the signal generator producing the 45 c.p.s. field sweep signal and was recorded by an Esterline-Angus recorder.

The samples used in these experiments were either saturated solutions or concentrated acids. Approximately one c.c. of the sample was placed in a $\frac{1}{2}$" diameter test tube which in turn was placed in the inductance coil of the super-regenerative oscillator tank circuit. The oscillator was so located in the magnet gap that the sample would occupy that portion of the field found by careful testing to be most homogeneous.

The tank circuit of the oscillator was tuned to a frequency somewhere in the range between 1 to 10 megacycles per second, depending upon the nuclear g-factor of the unknown reported by molecular beam experimenters and the magnetic field strength desired to be used for the experiment. This frequency could, after once being set by appropriate choice of fixed capacitance condensers, be varied within narrow limits by means of a small variable condenser in the tank circuit. In most cases the small variation in oscillator frequency permitted by the tuning condenser would not cover the frequency range required for comparison of the resonance frequency for the sample of undetermined g-factor with that of the known sample. This consideration required that a second set of fixed capacitance condensers be installed in the

pole face oscillator with necessary switching arrangements to permit rapid changing between the two frequency ranges.

The alternating quench voltage applied to the grid of the oscillator triode was variously set from 2.0 to 9.0 kilocycles and from 1.5 to 3.0 volts. These and other parameters were varied both in attempts to locate the resonance signal and, after the signal was found, to obtain a sharp, accurately measurable signal pattern. Other parameters varied for the same purpose were the magnetic field modulation strength, oscillator plate voltage, and the concentration and type of chemical compound used for the sample.

The signal from the pole face oscillator was sent through a narrow band amplifier tuned to filter out all alternating current components except the 45 cycles per second of the magnetic field modulation. The output of the narrow band amplifier was sent both to an oscilloscope and to a phase-detector. The output of this latter instrument was in turn passed to an Exterline-Angus recorder.

At resonance, corresponding to the condition

$$h\nu_0 = g_I \mu_n H,$$

the amplitude of oscillation of the super-regenerative oscillator is altered because of the previously discussed resonance phenomena. This amplitude change was observed visually on the oscilloscope trace and the time derivative of this change in amplitude of oscillation was recorded by the Esterline-Angus recorder from the output of the phase detector.

After resonance was observed and operating parameters were properly adjusted to obtain a signal suitable for measuring, the clock-

drive mechanism operating the potentiometer of the magnet current regulator was stopped and the field current regulator was allowed to maintain a constant current to the magnet field coils. The center of the resonance spectrum of the sample to be measured was accurately located by tuning the variable condenser in the pole face oscillator circuit and then measuring the oscillator frequency using the heterodyne frequency meter shown in Figure 3. With the magnet current still maintained constant, a sample of known value of nuclear g-factor was substituted for the unknown in the pole face oscillator and the oscillator frequency varied until resonance was obtained for this standard sample. With g_x and g_s denoting respectively the nuclear g-factors of the nucleus being studied and the nucleus used for standard, and ν_x and ν_s denoting their frequencies in the same magnetic field, g_x was obtained from the relation

$$g_x = g_s \frac{\nu_x}{\nu_s}.$$

Several pairs of such readings commonly were obtained and the average of their ratios was taken as the value for the measurement. Probable errors of measurements were computed according to customary procedures.

(b) Modulation Improvements

The magnetic field modulation for the initial period of this report was achieved by means of only a single sweep coil clamped on one pole piece. It was wound with about 2500 turns of 32 gauge wire and was supplied with 45 cycle voltage produced by an audio signal generator and amplifier. Sweep coil voltage was varied ordinarily in the range from 20 to 60 volts as necessary to obtain optimum

resonance signals.

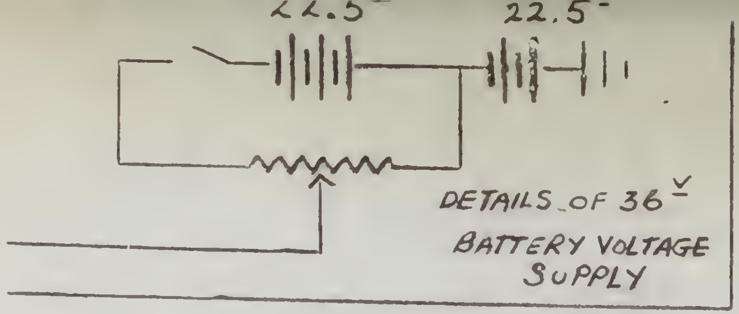
After the measurements on Indium^{115} were completed, tests were made using a small induction coil and an a.c. voltmeter to ascertain whether field modulation was appreciably constant over the region occupied by the sample in the gap between the pole faces. Although the small coil had a width equal to almost half of the pole face gap, a decrease in induced voltage of approximately fifty per cent was noted when the coil was held against the center of the face of the pole piece on which the sweep coil was mounted and from there moved parallel with the magnet axis to a position against the opposite pole face.

As a result of these tests it was decided to mount a similar coil on the second pole piece and to construct a push-pull amplifier to provide the sweep field power for both coils. The circuit diagram of this amplifier is shown in Figure 6. The new coil was over-wound by a few hundred turns to allow margin for proper balancing of the two coils. After the new amplifier and sweep coil were installed and the sweep field balanced by removing a certain number of excess turns from the newer coil, the variation in voltage induced in the probe coil was reduced to five per cent or less.

(c) Homogenizing of the Magnetic Field

The preparatory work attempted by the writer prior to making any actual measurements was performed using 5½" diameter pole pieces. Prior to that it had been the custom to fill the test tubes with sample solutions to a height of about two and one-half

Push-Pull Sweep Amplifier



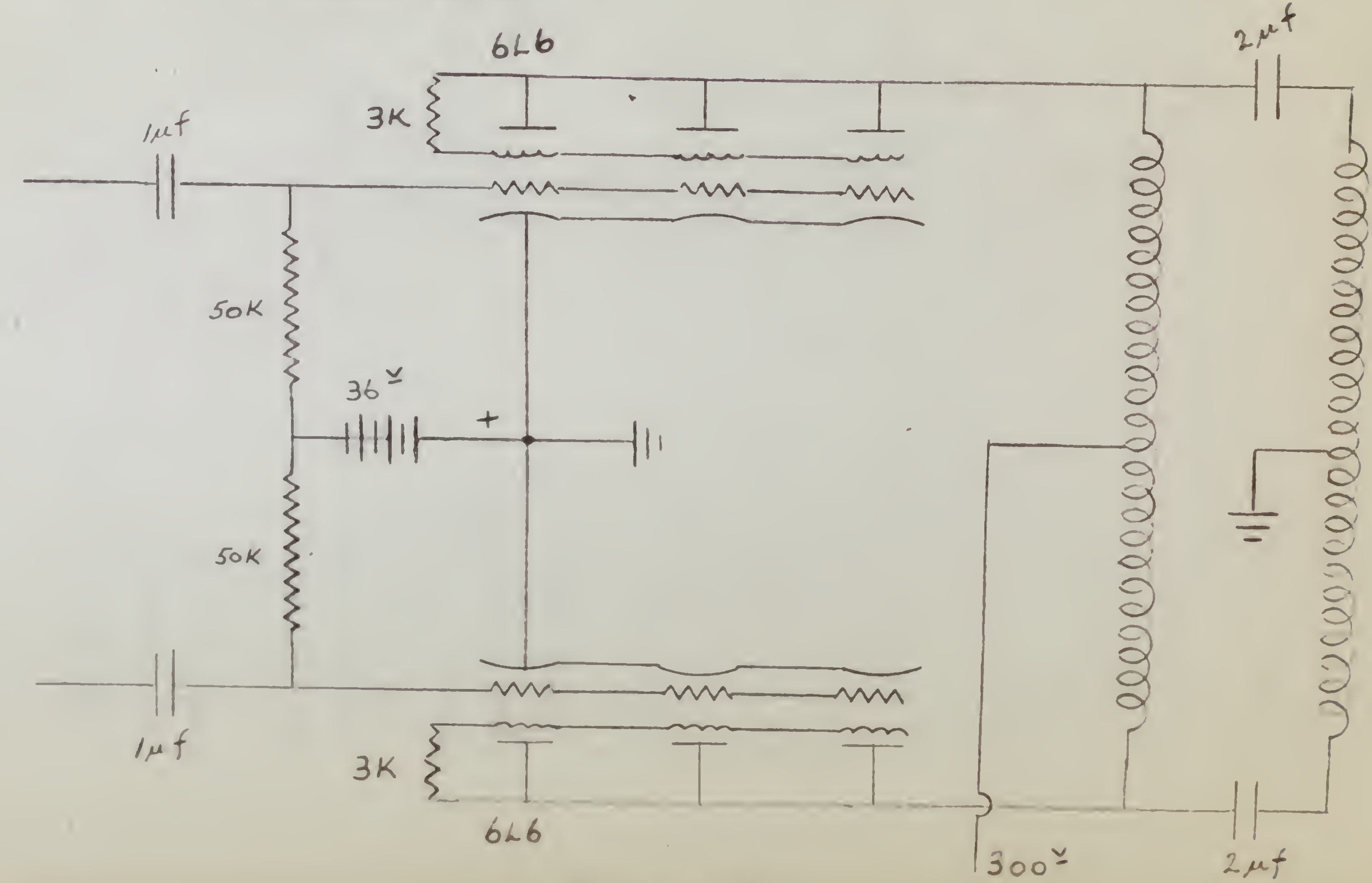


FIGURE 6.

inches. Utilizing a relatively large amount of sample in this manner ordinarily gave a strong resonance signal, but the center of the resonance pattern usually was lacking in the degree of sharpness desired for maximum accuracy of measurement.

It appeared possible that lack of desirably sharp resonance signals might be attributable in part to inhomogeneity of the magnetic field over the volume occupied by the sample. To check this assumption, test runs were made using smaller amounts of the sample solution. This procedure immediately produced sharper resonance patterns. After a number of tests it was ascertained that acceptable resonance signal strengths usually could be obtained using a depth of liquid in the tube ranging from 1.0 to 1.5 cm. Appreciably less than this amount for many nuclides frequently gave an undesirably weak signal. Exceptions to this last statement were the proton and the deuteron, both of which were found to give strong signals from only a few drops of H_2O or D_2O, respectively.

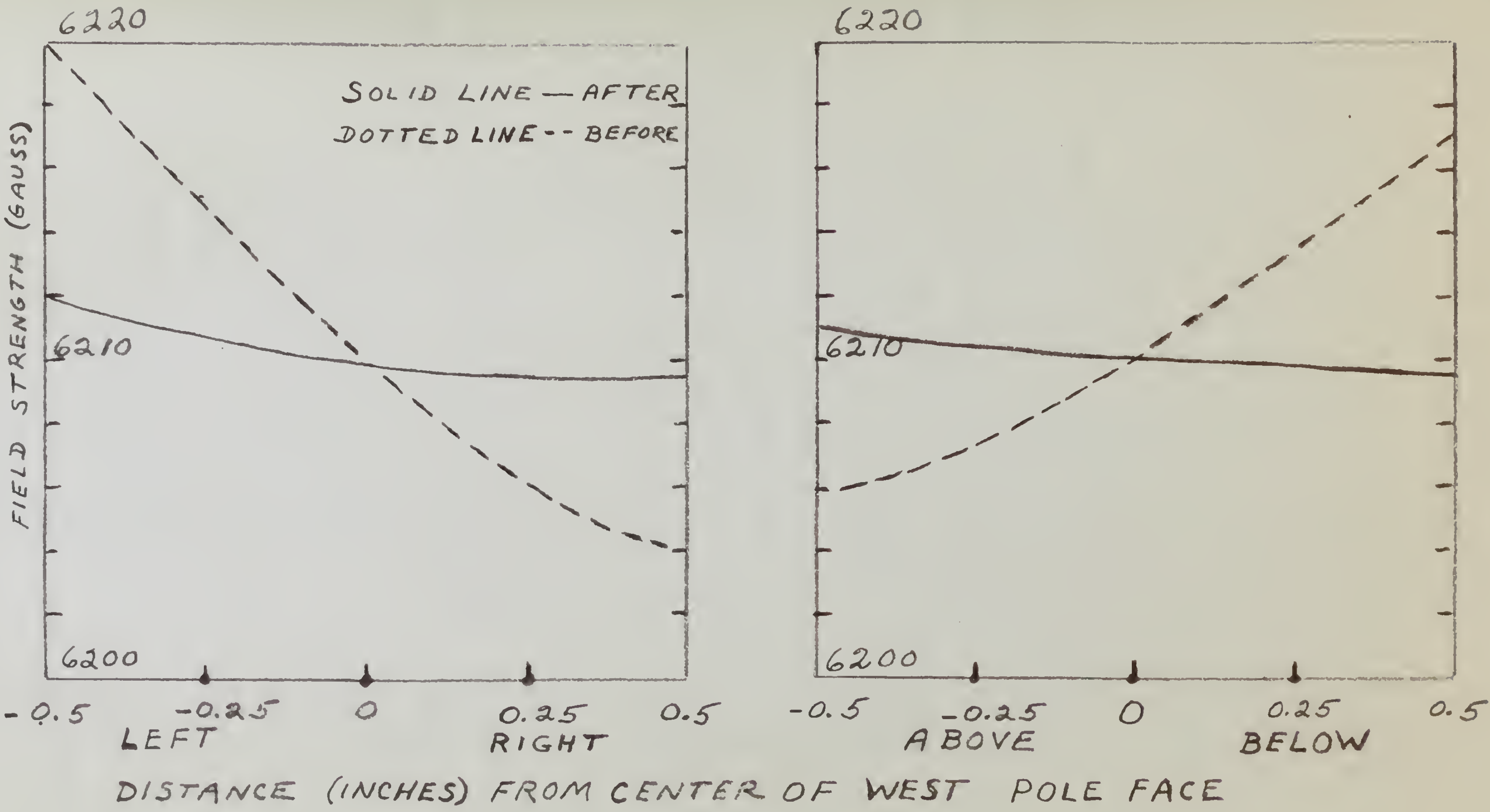
In a further attempt to achieve better field homogeneity, the 3½" pole pieces were replaced by others of 6" diameter. This change also produced an improvement in the sharpness of the resonance pattern but carried with it the slight disadvantage of placing a lower limit on the maximum field strength obtainable. This limitation was inconvenient upon occasions but was never found to be unacceptably restrictive.

In spite of the fact that the 6" pole pieces gave better resonance patterns it appeared probably that an additional improvement in field homogeneity could be obtained by removing a few slight burrs from the

pole faces. Further investigation into possible causes of field inhomogeneity also revealed that neither pole face was acceptably flat nor were the two faces sufficiently parallel. To correct all except the last of these deficiencies each pole face was carefully re-machined to make it as flat as possible and then was ground and polished by hand using emery dust and jeweler's rouge as grinding compounds. These efforts eventually produced reasonably flat and smooth pole face surfaces.

After the pole pieces were reinstalled a test was made to determine the most homogeneous part of the magnetic field for sample occupancy. For this check a small probe oscillator was employed using a sample of only some five or six drops of water from which to obtain proton resonance. Initial results indicated that the magnetic field even near the center of the pole faces varied as much or more than five gauss when the proton probe was moved a half inch in practically any direction. The pattern of variation indicated however, that shimming one side of one of the pole pieces might produce beneficial results. A long process of trial and error shimming using thin non-magnetic shims eventually produced a field which varied appreciably less than one gauss over a region sufficiently large to accomodate the desired height of liquid in a $\frac{1}{8}$" diameter test tube.

The deviation of the magnetic field from true homogeneity both before and after the remedial efforts described above is indicated in the graphs of Figure 7.



RESULTS OF HOMOGENIZING THE MAGNETIC FIELD
FIGURE 7.

(d) Results of Improvements

The results obtained from efforts spent to homogenize the magnetic field are well illustrated by differences appearing in the "before and after" resonance signals traced by the Esterline-Angus recorder. The typical "before" trace of a sharp resonance pattern appeared as an uninterrupted series of slow swings of the recorder pen in alternate directions. Pronounced hesitations in the movements of the pen occurred in this type of pattern only at each side limit of the recorder paper when the amplitude of the resonance signal exceeded the maximum capability of the recorder. A typical pattern of this type obtained from Indium^{115} is shown in Figure 8. In this pattern one might estimate that there are three resonance signals corresponding to the central signal and one pair of side-band resonances. Even so, there is nothing in the pattern which can be chosen as serving to separate the three theoretically distinct resonance signals.

The series of resonance signals in this pattern would be seen on the oscilloscope in the following sequence. Upon reaching the first resonance band the small fluctuations of the oscilloscope caused by random noise would be replaced by a steady buildup to a sustained peak of large amplitude followed presently by a steady decrease to a sustained valley of similar amplitude. This peak-valley would be twice repeated by the signal recorded in Figure 8. The trace on the oscilloscope in moving from valley to peak and vice-versa would do so without any significant hesitation as the trace crossed the horizontal axis. At the conclusion of the signal

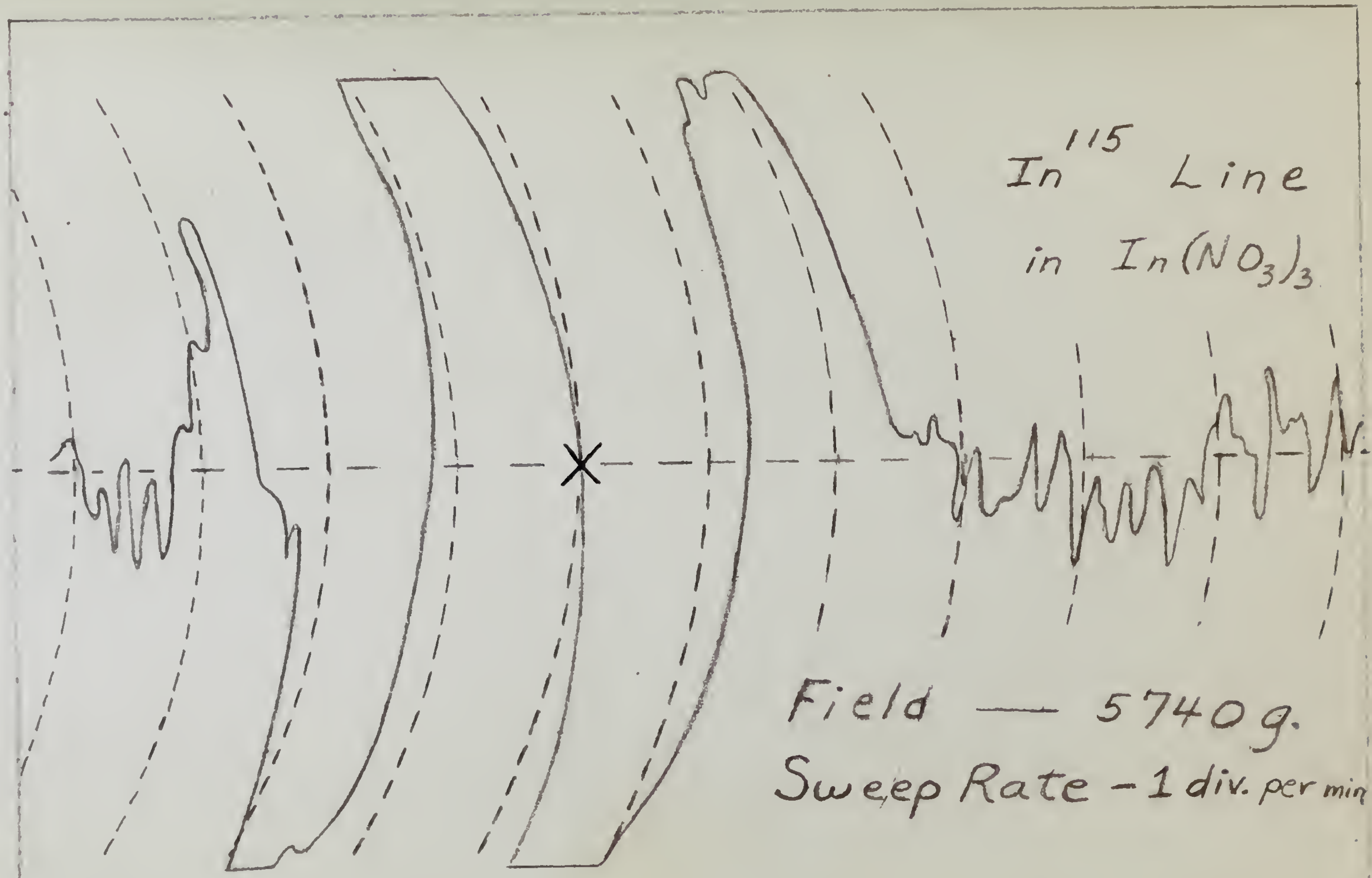


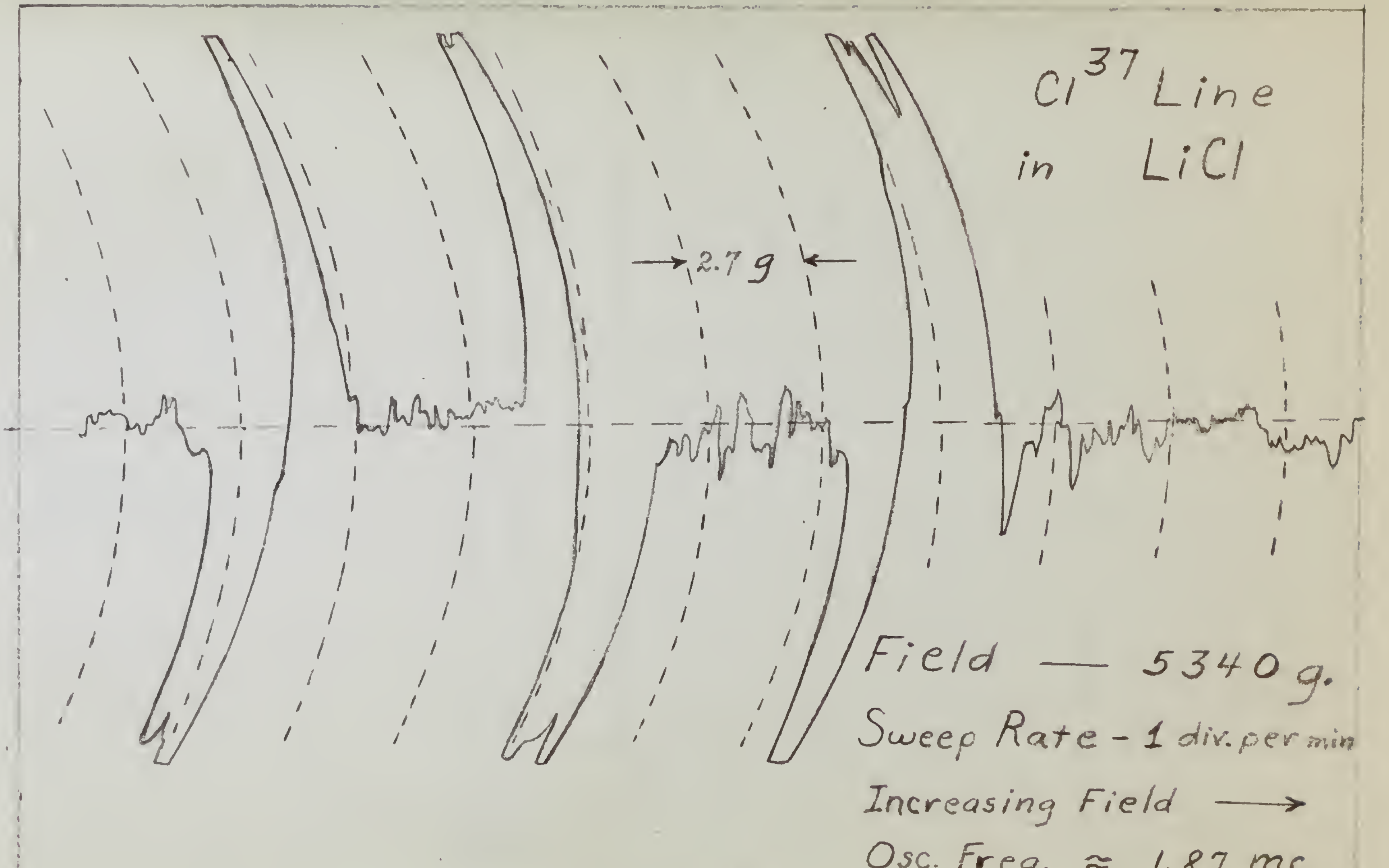
FIGURE 8.

the oscilloscope would again display random noise effects.

A different resonance pattern representative of the sharp "after" type is shown in Figure 9. In this pattern obtained from Chlorine[37] it may be seen that the central resonance signal is clearly separated from each of the two signals of the one sideband pair which are in evidence. This separation appeared on the oscilloscope as a return of random noise fluctuations for a period of approximately one and a half minutes between the separate resonance signals. It may also be noticed that the width of each signal is narrower, a feature surely attributable to better field homogeneity.

Another new feature which has not yet been adequately explained is the following phenomenon to be seen in Figure 9. Observing the direction of the initial excursions of the recorder pen in each of the three separate resonance signals one notices in the right hand signal that the first excursion is toward the top of the paper; that of the central signal is toward the bottom, while that of the left-hand signal is again upward. This alternation of direction of intial swings of the recorder pen is not confined merely to first order sidebands but has been noticed in all side band resonance signals observed since the field homogeneity was improved.

It will be recalled that the Esterline-Angus recorder receives its input from the super-regenerative oscillator through a narrow band amplifier and a phase detector. No conclusion has yet been reached, why the resonance signal as detected by this arrangement should behave in the above manner. Studies, however, are being



Cl^{37} Line
in LiCl

Field — 5340 g.
Sweep Rate - 1 div. per min
Increasing Field ⟶
Osc. Freq. ≈ 1.87 mc.

FIGURE 9.

continued by other personnel to attempt to explain this phenomenon.

A definite advantage in one phase of making frequency measurements on resonance signals accrued from the work on the pole pieces. In the older type of pattern one was forced to choose the point in the pattern representing the center of symmetry. It can be seen in Figure 8 that only the point indicated by X satisfies this condition. For an accurate measurement to be obtained it was essential that the swing of the pen across the region of the paper near the X occur without any hesitation. Unfortunately, in practice this condition is difficult to obtain.

With the newer resonance pattern in which each side band resonance signal was clearly distinguishable it was discovered to be possible to shift to a sharp side band if the central resonance signal happened not to be sharp. Having shifted to a side band to obtain frequency measurements one then only had to apply the correction

$$n\nu_{gu}$$

to the observed side band resonance frequency ν_s to obtain the desired but unobserved central resonance frequency ν_0. The relation employed for this purpose is

$$\nu_s = \nu_0 + n\nu_{gu},$$

in which n, the order of the side band, is given the appropriate algebraic sign.

Measurements obtained from side bands were in no manner less accurate than those obtained from the central resonance signal.

Although most nuclei seldom exhibited more than two or three pairs of side bands in a resonance pattern, at least ten such pairs

of side bands have been observed frequently in resonance signals produced by the deuteron in the homogenized magnetic field. The upper limit on n for the deuteron can not be stated with accuracy because, in the interest of economy of time and ease of measurement, efforts were usually exerted to limit the number of side bands in a pattern consistent with obtaining the desired degree of sharpness in the signal.

IV. EXPERIMENTAL RESULTS

The measurements to be described in this section were obtained by comparing in the same magnetic field the resonance frequency of each unknown with that of a known sample used as a standard. The magnetic moment of the unknown sample was then obtained from the relation

$$\mu_x = g_x I_x = g_p \left(\frac{\nu_x}{\nu_s}\right)\left(\frac{\nu_s}{\nu_p}\right) I_x$$

In each case the ratio $(\nu_{\text{standard}}/\nu_{\text{proton}})$ has been obtained from recent results published by other investigators. The value used for the g-factor of the proton was

$$g_p = 5.58536 \pm 0.00012,$$

obtained from the value $\mu_p = (2.79268 \pm 0.00006)\mu_n$.[5]

The probable error indicated in each table for the ratio $(\nu_{\text{unknown}}/\nu_{\text{standard}})$ is a measure of the internal consistency of the data. Since the existence of systematic errors as large as one part in twenty thousand can not be excluded, the probable error of the magnetic moment has been computed using the systematic error if that error exceeded the probable error of the ratio $(\nu_{\text{unknown}}/\nu_{\text{standard}})$.

The results here have not been corrected for diamagnetic effects of the extra-nuclear electrons nor for the slight paramagnetic effect of a magnetic catalyst in those cases in which it is indicated that such a catalyst was used.

(a) Indium^{115}

The measurement of the nuclear magnetic moment of In^{115} was obtained from one c.c. of a saturated solution of $In(NO_3)_3$ in 30% HNO_3 to which had been added a small amount of $Mn(NO_3)_2$ as a catalyst. The standard used for comparison was Sc^{45} in one c.c. of a saturated solution of $ScCl_3$. The value for the ratio

$$\frac{\nu_{In^{115}}}{\nu_{Sc^{45}}} = 0.242939 \pm 0.00003$$

is that due to Hunten.[26] The data pertaining to this measurement are summarized in Table 1.

Table 1.

Data for Indium^{115}

Unknown Nuclide	Indium^{115}
Standard Nuclide	Scandium^{45}
$\frac{\nu_{In^{115}}}{\nu_{Sc^{45}}}$	0.902292 ± 0.00005
$\frac{\nu_{Sc^{45}}}{\nu_{H^1}}$	0.242939 ± 0.000003
$\frac{\nu_{In^{115}}}{\nu_{H^1}}$	0.219202 ± 0.000012
$g_I(In^{115})$	1.22452 ± 0.00012
$\mu_I(In^{115})$	(5.50945 ± 0.00015)

---

[26] Hunten, D.M. Phys. Rev. 78, 806 (1950).

(b) Chlorine^{37}

The measurement of the nuclear magnetic moment of Cl^{37} presented in Table 2 was obtained using a saturated solution of approximately one c.c. of LiCl to which had been added a small amount of $MnCl_2$ as a magnetic catalyst. The deuteron resonance frequency used as a standard was obtained from a one c.c. sample of D_2O. The value of the ratio

Table 2.

Data for Chlorine^{37}

Unknown Nuclide	Chlorine^{37}
Standard Nuclide	Deuteron
$\frac{\nu_{Cl^{37}}}{\nu_{D^2}}$	0.531632 ± 0.000036
$\frac{\nu_{D^2}}{\nu_{H^1}}$	0.153506
$\frac{\nu_{Cl^{37}}}{\nu_{H^1}}$	0.0816087 ± 0.0000054
$g_I(Cl^{37})$	0.455848 ± 0.000032
$\mu_I(Cl^{37})$	0.683722 ± 0.000048

$\frac{\nu_{D^2}}{\nu_{H^1}} = 0.153506$ is taken as being accurate to six significant figures.[27]

---

[27] Smaller, B., Yasaitis, E., and Anderson, H.L. *Phys. Rev.* 80, 137(A) (1950).

(c) Nitrogen^{14}

The measurement for N^{14} summarized in Table 3 was obtained using approximately one c.c. of concentrated HNO_3. Rb^{85} in a one c.c. saturated solution of RbCl was used as the standard. The ratio $(\nu_{Rb^{85}}/\nu_{H^1})$ was obtained from results by Yasaitis and Smaller in which the ratio was given as[28]

$$\frac{\nu_{H^1}}{\nu_{Rb^{85}}} = 10.357105 \pm 0.00001.$$

Table 3.

Data for Nitrogen^{14}

Unknown Nuclide	Nitrogen^{14}
Standard Nuclide	Rubidium^{85}
$\frac{\nu_{N^{14}}}{\nu_{Rb^{85}}}$	0.748425 ± 0.000019
$\frac{\nu_{Rb^{85}}}{\nu_{H^1}}$	0.096552 ± 0.0000001
$\frac{\nu_{N^{14}}}{\nu_{H^1}}$	0.072262 ± 0.000002
$g_I(N^{14})$	0.40361 ± 0.00002
$\mu_I(N^{14})$	0.40361 ± 0.00002

---

[28] Yasaitis, E., and Smaller, B. Phys. Rev. 82, 750 (1951).

V. DISCUSSION OF RESULTS

During the time which has elapsed since the measurements listed in Section IV. were obtained, another member of the laboratory, Dr. Yu Ting, has obtained measurements of the nuclear magnetic moments of both Cl^{35} and In^{113}. In the measurement of Cl^{35} the standard used was Cl^{37} in the same sample. In the case of In^{113}, In^{115} was used as a standard. The In^{113} was contained in enriched proportions in a sample supplied on loan by the U.S. Atomic Energy Commission. The results tabulated in Tables 4 and 5 include the results obtained both by Dr. Ting and by the writer. In addition there are listed in Table 4 for the purpose of later discussion the experimental values of the quadrupole moments for the various nuclides as well as the nuclear magnetic moments of In^{113} and In^{115} obtained by the atomic beam method.

It will be noticed in Table 4 that the atomic beam values of the magnetic moments of both isotopes of Indium are less than the values obtained by magnetic resonance methods. These differences are much too large to be accounted for within the probable errors of the experiments. Similar discrepancies have been noted[29,30,31] for the isotopes Ga^{69} and Ga^{71}.

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[29] Taub, H., and Kusch, P. Phys. Rev. 75, 1481 (1949).

[30] Pound, R.V. Phys. Rev. 73, 1112 (1948).

[31] Kusch, P. Phys. Rev. 73, 584 (1948).

Table 4.

Data on μ_I* and Q

Nuclide	Z	A-Z	I	μ_I(Mag. Res.)	μ_I(Atomic Beam)	Q ($10^{-24}cm^2$)	
Cl^{35}	17	18	3/2	0.820896 μ_n		-0.0795 ± 5	a.
Cl^{37}	17	20	3/2	0.683722		-0.0621 ± 5	a.
In^{113}	49	64	9/2	5.49507	5.486 a.	1.144	b.
In^{115}	49	66	9/2	5.50945	5.500 a.	1.161	b.
N^{14}	7	7	1	0.40361		0.02	a.

*Values for μ_I by magnetic resonance method were obtained at this institution.

a. Mack, J.E. Revs. Med. Phys. 22, 64 (1950).

b. Mann, A.K., and Kusch, P. Phys. Rev. 77, 427 (1950).

Table 5.

Comparison of Results with Those Obtained Elsewhere

Nuclide	μ_I (Mag. Res. from Table 4.)	μ_I (Proctor and Yu[32])
Cl^{35}	(0.820896 ± 0.000050)μ_n	(0.8211 ± 0.0001) μ_n
Cl^{37}	0.683722 ± 0.000048	0.6835 ± 0.0001
In^{113}	5.49507 ± 0.00020	5.4972 ± 0.0010
In^{115}	5.50945 ± 0.00015	5.5088 ± 0.0010
N^{14}	0.40361 ± 0.00002	0.40369 ± 0.00006

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[32] Proctor, W.G., and Yu, F.C. Phys. Rev. 81, 20 (1951).

Foley[33] has investigated a suggestion by Townes that partial decoupling of the L and S vectors in the $^2P_{\frac{1}{2}}$ state in the applied magnetic field of the atomic beam apparatus might affect the hyperfine interaction in such a way that the nuclear moment appears to be altered. The results obtained by Foley and his co-workers using a non-relativistic one-electron wave function in developing an expression for the energy perturbation of the magnetic levels of the $^2P_{\frac{1}{2}}$ state have indicated that the effect does not quite account for the entire discrepancy observed. Foley has further pointed out that the ground states of such atoms as gallium, indium and thallium are subject to configuration interaction mixing with some excited states of the outer three electrons, which affects strongly the hyperfine splittings of the $^2P_{3/2}$ states. This fact might possibly account for the remaining discrepancy.

The values of the results obtained by the writer and also those by Dr. Ting as listed in Table 5 are seen to agree with those obtained by Proctor and Yu at Stanford within one digit at the fourth significant figure within the limits of probable errors. Results obtained by other investigators generally come within limits as close or smaller than this.

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[33] Foley, H.M. *Phys. Rev.* *80*, 288 (1950).

(a) Shell Theory

The statement was made in Section I that no theory at present is capable of predicting magnetic moments of nuclides having non-zero spin. There is a model of the nucleus due to Schmidt,[34,35,36] however, which attempts to explain nuclear magnetic moments by assuming that the nuclear spin I in an odd-even or even-odd nucleus results from the motion of the one unpaired nucleon. The sum of the orbital and spin angular momenta of all other nuclear particles is assumed to be zero. For this model we have

$$\underline{I} = \underline{L}_K + \underline{S}_K \qquad \text{AND}$$

$$\mu_I = \frac{eh}{4\pi Mc}\left(g_L \underline{L}_K + g_S \underline{S}_K\right),$$

where g_L and g_S are gyromagnetic ratios with values

$$g_L = 1 \qquad \text{and}$$

$$g_S = g_P = 1$$

if the odd particle is a proton, or

$$g_L = 0 \qquad \text{and}$$

$$g_S = g_n = (-)3.82$$

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[34] Schmidt, T. Zeits. f. Physik 106, 358 (1937).

[35] Fermi, E. Nuclear Physics. Chicago: The University of Chicago Press, 1950, p. 14.

[36] Blatt, J.M., and Weisskopf, V.F. Theoretical Nuclear Physics. New York: John Wiley & Sons, 1952, pp. 38-39, 767-774.

if the odd particle is a neutron. Since by assumption only the unpaired particle contributes to the nuclear spin I, the result must follow that $S = \frac{1}{2}$ or $S = -\frac{1}{2}$, so that $L = I + S$ must have either the value $L = I + \frac{1}{2}$ or $L = I - \frac{1}{2}$. Further, the parity of the state of the nucleus must be either odd or even, but cannot be both, so the orbital angular momentum ℓ of the unpaired nucleon must be either $\ell = I + \frac{1}{2}$ or $\ell = I - \frac{1}{2}$. With these assumptions it is possible to derive the Schmidt equations[37] in the following forms:

$$\mu_I = I + 2.29 \qquad \text{odd proton, } I = \ell + \tfrac{1}{2},$$

$$\mu_I = \frac{I^2 - 1.29I}{I+1} \qquad \text{odd proton, } I = \ell - \tfrac{1}{2},$$

$$\mu_I = -1.91 \qquad \text{odd neutron, } I = \ell + \tfrac{1}{2}, \text{ and}$$

$$\mu_I = \frac{1.91I}{I+1} \qquad \text{odd neutron, } I = \ell - \tfrac{1}{2}.$$

Since the assumptions require that $I = \ell \pm \frac{1}{2}$, the Schmidt equations thus will yield two values for the predicted μ_I of any particular nuclide. It is found in practice that nuclear magnetic moments observed to date will, with only a few exceptions, lie between the two values yielded by these equations. Those observed values which are exceptions fall outside of these limits only by comparatively small amounts.

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[37] Fermi, E. Nuclear Physics. Chicago: The University of Chicago Press, 1950, pp. 19-21.

Unfortunately, the difference between the two Schmidt limits extends over ranges covering differences from two to four nuclear magnetons. This lack of preciseness seriously limits the usefulness of the theory.

By adopting some of the results contained in the theory of closed shells in nuclei as proposed by Mayer[36] and others it is possible to limit the choice of ℓ for the odd particle to only one value. By this theory it is found that $\ell = 2$ for both Cl^{35} and Cl^{37}; for In^{113} and In^{115}, $\ell = 4$.

The two values of the nuclear magnetic moments predicted by the Schmidt equations for Cl^{35} and Cl^{37}, for In^{113} and In^{115}, and for N^{14} together with the observed values are listed in Table 6. The values obtained by using the nuclear shell theory results for ℓ with the appropriate Schmidt equation to obtain a single value for each μ_I are indicated by asterisks.

The examples in Table 6 are too limited in number to indicate the proper degree of merit which should be attached to the above method of attempting to predict values of μ_I. That this procedure leaves much to be desired would be seen, however, if one were to consider the cases which should be predicted most accurately. These cases consist of nuclides with an even number of neutrons and one more proton than the number required to complete the filling of a shell. It would seem in these cases that fairly close agreement should be obtained between theory and experiment, for the filled "core" of protons should be spherically symmetric and the magnetic moment should be due only to the one additional proton. But even

Table 6.

Predictions of μ_I From Schmidt Equations and Shell Theory

Nuclide	Spin	ℓ	μ_I(Schmidt Model)		μ_I(Observed)	Diff. (S.m.-obs.)
		(Shell Theory)	Lower	Higher		
$_{17}Cl^{35}$	3/2	2	0.126*	3.79	0.820896	-0.695
$_{17}Cl^{37}$	3/2	2	0.126*	3.79	0.683722	-0.458
$_{49}In^{113}$	9/2	4	2.60	6.79*	5.49507	+1.29
$_{49}In^{115}$	9/2	4	2.60	6.79*	5.50945	+1.28
$_{7}N^{14}$	1	(- - - -Not Applicable- - -)			0.40361	

* indicates value obtained using ℓ from nuclear shell theory.

in these cases the predictions are found to be no more accurate than those in many cases less favorable.[36]

In^{113} and In^{115}, two of the isotopes represented in Table 6, belong to a class of isotopes having identical spins but having N = A - Z differing by two, where N is the number of neutrons in the nuclide. Frequently, pairs or triplets of isotopes of this type have almost identical magnetic moments. Examples of these are Ag^{107} and Ag^{109}; Cs^{133}, Cs^{135}, and Cs^{137}; Tl^{203} and Tl^{205}; and In^{113} and In^{115}. This indicates that the protons, which according to Schmidt theory are responsible for the magnetic moments of all of these even-neutron nuclei, do not have their arrangement very greatly disturbed when two more neutrons are added to the nucleus. In^{113} and In^{115} also have very similar quadrupole moments and isomeric levels.[36]

(b) Relationships Between μ_I and Q

For nuclei containing closed proton shells plus or minus one, the nuclear shell model not only predicts the sign of the quadrupole moment, but if the state of this odd proton can be ascertained from the nuclear spin and the magnetic moment, it is possible to arrive at a fairly good value for the quadrupole moment.[38] The nuclear shell model also leads to other less quantitative conclusions concerning quadrupole moments.

Townes and his collaborators[38] have proposed a simple model of the nucleus based on nuclear shell considerations which leads to the proper behavior of known quadrupole moments but leaves the predictions of some of their magnitudes appreciably in error. This model is characterized by the following:

1. Neutrons and protons fit into single particle levels in a scheme similar to those proposed for correlating spins, thus producing what may be called proton and neutron shells.

2. Proton and neutron shells tend to be oriented or polarized to allow maximum overlap between proton and neutron distributions.

This model is stated by its originators to lead to the following conclusions:

A. For an odd-proton nucleus, the quadrupole moment is primarily dependent on the number of protons P and can be written $Q_{p\ odd} = Q_p(P)$, in which Q_p is always positive immediately before,

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[38] Townes, C.H., Foley, H.M., and Low, W. Phys. Rev. 76, 1415 (1949).

and always negative immediately after a shell is filled.

B. For an odd-neutron nucleus, the magnitude of the quadrupole moment depends on the number of protons, but its sign is determined by the number of neutrons, N, being given by $(Q_p(N)/|Q_n(N)|)$. In this expression $Q_n(N)$ is the electric quadrupole moment which would be produced if they were protons. $Q_n(N)$ is very nearly the same function as $Q_p(N)$.

C. For odd-odd nuclei, estimation of quadrupole moments is more complex and depends on the way in which the angular momenta of the odd neutron and odd proton add. If these moments are essentially parallel, the quadrupole moment should be of the same sign and approximately the same magnitude as for a similar odd-proton nucleus. If the neutron and proton angular momenta are not essentially parallel, the quadrupole moment magnitude should be considerably reduced.

Generally it may be said that the nuclear shell model does not give correct values of Q. This is in contrast to the nuclear magnetic moments, for which an appropriate admixture of states of one nucleon can account for the magnetic moment of the nucleus, although this procedure does not appear very plausible by even the presently known facts about the nucleus. Large quadrupole moments demand an appreciable contribution from the protons which, according to the shell model, are in closed shells. For this contribution to be realized, it appears that angular momentum would have to be shared between the protons of the incomplete and of the filled shells. This polarization and the large asymmetry of distribution of nucleons is not entirely compatible with the single-particle central field concept

which forms the foundation of shell theory.[58]

Hence it may be said that there is no well understood relationship between μ_I and Q. Both are dependent upon spin, and both are functions of the number of protons and neutrons in a nucleus. To date no theory has been successful in establishing any important relationship between these two quantities.

ACKNOWLEDGMENTS

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The writer wishes to express his sincerest appreciation for the extensive advice and encouragement given him in the above work by his adviser, Professor Dudley Williams, and to Dr. Yu Ting for his extremely valuable assistance in all phases of the work covered in this thesis.

Thesis
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